The Nuclear Alchemy Gamble

An Assessment of Transmutation as a Nuclear Waste Management Strategy

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August 25, 2000

(May 12, 2005 web-posted version with modifications to Appendix B)

Acknowledgments

We would like to thank Dr. Edwin Lyman, David Lochbaum, Greg Mello, Dr. Lawrence Lidsky, Hideyuki Ban, Dr. Yukio Yamaguchi, Monique Sené, and Jean-Pierre Morichaud for their review of one or more drafts of this report. Of course, the authors are solely responsible for the content of the report, its conclusions and recommendations, and any omissions or errors that remain. We would also like to thank IEER staff members Lois Chalmers, Diana Kohn, Betsy Thurlow-Shields, Michele Boyd and Arjun Makhijani for their comments and assistance.

Research for this report was conducted as part of IEER's program: *The Road to Enduring and Complete Nuclear Disarmament: A Technical Outreach Project on Nuclear Weapons and Related Energy Issues.* We gratefully acknowledge the W. Alton Jones Foundation, John D. and Catherine T. MacArthur Foundation, the C.S. Fund, and the HKH Foundation for their generous support of this program. Outreach for this report will also be conducted as part of IEER's *Project to Provide Technical Assistance to Grassroots Groups on Nuclear Issues*, which is made possible by the support of the John Merck Fund, Ploughshares Fund, Public Welfare Foundation, Town Creek Foundation, and the Unitarian Universalist Veatch Program at Shelter Rock. We would also like to thank our individual donors and general support funding of the Beldon II Fund, Stewart R. Mott Charitable Trust, Rockefeller Financial Services, and Turner Foundation.

"Research on partitioning and transmutation is rather seductive to all of us. It requires new reprocessing techniques, new fuel developments, additional nuclear data, new reactors and irradiation facilities, new waste treatment and disposal concepts, and specific safety studies. The global nuclear scientific and engineering community is challenged by this opportunity."
"Everybody realizes however that this voyage to the promised land will pass a desert with a lot of mountains and that we are not so sure that the horizon will be as bright as one can hope."

---Paul Govaerts, SCK-CEN (Belgian Nuclear Research Center). "Welcome Address" to the Fifth International Information and Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Mol, Belgium, 25-27 November 1998.

"The [transmutation] programme is expected to serve to revitalise the nuclear R&D in general, and also to attract capable young researchers dedicated to bringing the nuclear option into the 21st century in a healthy state."

---"OMEGA Programme: Partitioning and Transmutation R&D Programme of Japan," in Organisation for Economic Co-Operation and Development/Nuclear Energy Agency, *Actinide and Fission Product Partitioning and Transmutation: Status and Assessment Report*, Paris: OECD/NEA, 1999, page 253.

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Executive Summary

One of the biggest obstacles facing the nuclear industry is what to do with the nuclear waste generated in the form of spent fuel discharged from commercial reactors or in the form of high-level waste originating from the extraction of plutonium from spent fuel.[1]

Most countries' preferred option for the isolation of nuclear waste from the public and the environment is to bury it underground in a deep geological repository. However, because the spent fuel and the high-level waste contain a number of radionuclides that have very long half-lives (thousands of years to millions of years) it is generally acknowledged that it is impossible to ensure the isolation of the waste for such long periods of time. Besides the likelihood of leakage of some long-lived radionuclides, it is also impossible to guarantee against human intrusion (intentional or inadvertent).

The extremely difficult questions regarding ensuring isolation of waste to a degree sufficient to prevent severe contamination of resources, notably water resources, has made the siting of repositories a controversial scientific and policy issue and has been at the center of much of the public concern and opposition to repositories. Further, the political expediency that has frequently accompanied the selection of sites for study has intensified this opposition. While programs for siting repositories for spent fuel and high level waste are in various stages in different parts of the world, these still face immense scientific hurdles and intense public opposition. In the United States, which has a 2010 target date for opening a repository, there are still no final environmental standards for the protection of the health of future generations and of the environment from the proposed repository at Yucca Mountain.[2]

The difficulties and questions associated with repository siting, notably the extremely long periods of isolation required, have caused some to view the transmutation of long-lived radionuclides into short-lived ones as a potential solution to the problem of radioactive waste management. Transmutation is done by inducing nuclear reactions of various types in the nuclei of long-lived radionuclides. The theory is that a transmutation program would transform the vexing problem of long-term isolation into a far less difficult one of storage for several decades or a few hundred years.

This *theoretical* promise has led proponents of transmutation to claim that it would greatly decrease the problems associated with long-term management of nuclear waste. Occasionally, they have even claimed that it might eliminate the need for a repository, though such claims have tended to recede as investigations into the practicalities of transmutation have progressed. At the same time, environmental, waste management,

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[1] There are over 400 nuclear power reactors currently operating worldwide. About 220,000 metric tons of spent fuel have been discharged from these reactors to date (the year 2000), and the number is increasing at a rate of about 10,000 metric tons per year. Almost 20 percent of the plutonium in this fuel has been extracted by reprocessing, while the rest is stored as spent fuel. See IAEA 1997b, p. 119, and Energy Information Administration, U.S. Department of Energy. *World Spent Fuel Discharges, Reference Case, 1999-2020*. http://www.eia.doe.gov/cneaf/nuclear/n_pwr_fc/data98/table10.html. For reprocessing data and estimates, see Albright, Berkhout, and Walker 1997, Chapter 6.

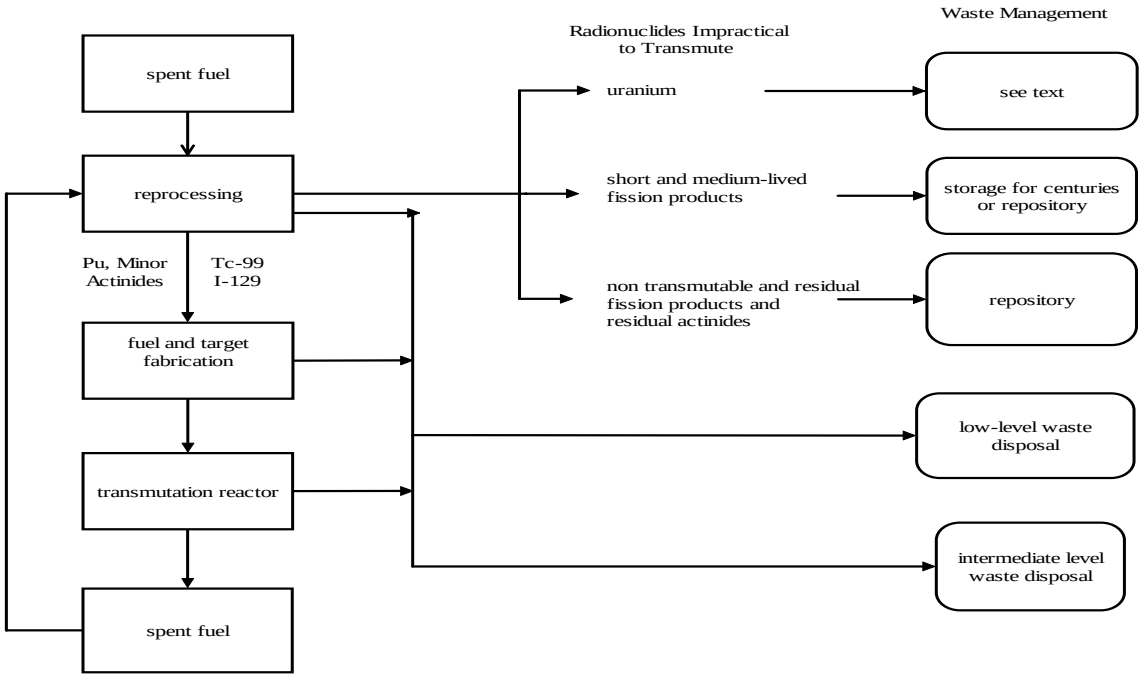
[2] See *Science for Democratic Action* vol. 7, no. 3 (May 1999) for more information about issues related to the long-term management of nuclear waste, particularly in the United States, and for just some of the evidence concerning Yucca Mountain's unsuitability as a repository location.

cost, and proliferation concerns have risen. In addition to its promise of a solution to the nuclear waste problem, some transmutation proponents have touted it as the only complete solution to the proliferation problems posed by plutonium. They argue that as long as plutonium remains, either in stockpiles of separated plutonium or in spent fuel that can be reprocessed to obtain separated plutonium, the proliferation risks will remain. Their solution is to use the plutonium as fuel in reactors even if this requires the separation of the plutonium and therefore an increase in proliferation risks over the short term.

Transmutation basics

Transmutation is the transformation of a radionuclide into another radionuclide, or into two or more radionuclides. Nuclear waste transmutation involves nuclear reactions that would occur in some form of nuclear reactor (thus producing electricity at the same time as transmuting the radionuclides).[3] A variety of reactor schemes have been proposed, but they all possess a common characteristic: a substantial amount of energy must be delivered to the nucleus of a long-lived radionuclide in order to induce a nuclear reaction that would convert it into a short-lived radionuclide or a stable element.

Figure 1: Stages of the Transmutation Process



[3] Reactors do not necessarily have to produce electricity. For instance, with one exception, none of the reactors used to produce plutonium for nuclear weapons in the United States produced electricity. However, the sale of electricity is the only way to recoup some of the high costs associated with transmutation. This requirement can create its own problems, however, by raising the reliability requirements of some transmutation systems so as to not disrupt electricity supply once it is operational (see section on accelerator reliability in Chapter IV).

The figure above shows the main components of an idealized transmutation system. A reprocessing plant is needed to sort out the candidate radionuclides slated for transmutation by separating certain long-lived radionuclides from the others. (In the context of transmutation, reprocessing is also called "separation" or "partitioning.") This allows the selective conversion of long-lived radionuclides into short-lived ones when they are irradiated in a reactor. Without reprocessing, the opposite kind of nuclear reactions would cause a counterproductive conversion of some short-lived radionuclides into long-lived ones.

The fabrication facility then manufactures the long-lived radionuclides into fuel and/or targets that are then sent to the transmutation facility, where the conversion of the nucleus actually takes place. The central component of a transmutation facility is a nuclear reactor. It may be a critical reactor, which is a self contained transmutation device, or a sub-critical reactor, which needs an outside source of neutrons to sustain a chain reaction.[4]

The neutron induced reactions in the reactor transmute the long-lived fission products into short-lived ones; they also fission the actinides, such as plutonium, creating new fission products. Most of these fission products are short-lived, but new long-lived fission products are also created (see below). The actinides, like uranium and plutonium, can also absorb neutrons, resulting in the creation of higher-mass actinides (see below). So plutonium and other transuranic radionuclides are actually being created in some portions of the fuel in transmutation devices, while in others they are being destroyed. Further, not all actinides can be transmuted before the nuclear reactor becomes very inefficient. Hence, a number of passes through the reprocessing, fuel fabrication, and reactor facilities are needed in order to transmute most long-lived radionuclides.

Transmutation of all long-lived radionuclides into short lived ones to a degree sufficient to obviate the need for a geologic repository is practically impossible. In particular, the transmutation of separated uranium, which constitutes about 94 percent of the weight of light water reactor spent fuel and which is very long-lived and generally contaminated with some fission products, would be counterproductive. The main transmutation route for almost all the uranium would be to convert uranium-238 (the dominant isotope) into plutonium-239. Hence, the complete transmutation of uranium-238 essentially requires the creation of a plutonium economy, which would be unsound whether viewed from an economic, environmental, or non-proliferation standpoint. Almost all the uranium must therefore be disposed of without transmutation as a matter of practical necessity. Other long-lived fission products as well as residual transuranic actinides would also need disposal. Hence, a repository, as well as other waste management and storage facilities would still be an essential part of transmutation schemes.

The merits of transmutation schemes and the difficulties associated with them become clearer if we understand some basics about the physics of transmutation.

[4] Accelerated protons hitting a target made of heavy metal, which produces neutrons through a nuclear reaction called spallation, would produce the supplemental neutrons.

The physics of transmutation

Two transmutation reactions are important for nuclear waste management: neutron capture and fission.[5] The goal is that long-lived radionuclides be transformed into short-lived radionuclides that then decay into stable isotopes.

To provide concrete examples, this section will discuss neutron capture by two long-lived fission products: iodine-129 and cesium-135. In addition we illustrate two reactions involving plutonium-239 transmutation.[6]

The absorption of a neutron by iodine-129 results in the production of short-lived I-130 and then in the stable isotope xenon-130.[7] Cesium-135 captures a neutron to become short-lived Cs-136, which decays into stable barium-136.[8] Hence, in these two cases, nuclear theory indicates that transmutation of these troublesome long-lived radionuclides into non-radioactive, stable ones is possible. However, as a practical matter only I-129 can actually be considered a candidate for transmutation. In the case of cesium-135, transmutation would first require the separation of this specific isotope from cesium-133, which is stable. This is because successive capture of neutrons by cesium-133 converts it first into Cs-134 (short-lived) and then into Cs-135, which is long-lived.[9] The cesium in spent fuel is a mixture of both Cs-133 and Cs-135 isotopes which cannot feasibly be separated, in part because the presence of the very radioactive Cs-137 isotope makes the handling and processing of the cesium extremely difficult, expensive, and dangerous. Thus, it is easy to see that the benefit of transmuting Cs-135 would be negated by the production of more Cs-135 from the neutron capture of Cs-133.

Some neutrons interactions with plutonium-239 result in fission while others result in the formation of plutonium-240 with a half-life of 6,500 years, which while shorter than the 24,000-year half-life of Pu-239, is evidently still very long. Successive neutron captures result in higher plutonium isotopes.[10]

This illustrates that transmutation nuclear reactions would need to be closely controlled so that there is an overall change from long-lived to short-lived radionuclides without a build up of new long-lived radionuclides.

Note also that neutron capture by plutonium-239 and -240 would not solve the problem of eliminating long-lived radionuclides even if all the plutonium were converted to short-lived plutonium-241. This is because plutonium-241 has an entire decay chain associated with it. It decays into americium-241, which has a half-life of 430 years. Amercium-241 in turn decays into neptunium-237, which has a half-life of over 2 million years. It is

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[5] Transmutation is also possible using photonuclear reactions, which use energetic photons to induce transmutation. Photonuclear transmutation schemes share many technical details with schemes discussed in this report and pose essentially the same major problems. However, phototransmutation is even less developed and would pose even greater research and development hurdles.

[6] Reactions are shown in the footnotes with half-lives shown in parentheses. n = neutron; e = beta particle; m = metastable (an excited state of the nucleus that does not decay immediately to the ground state). Half-lives are rounded to two significant figures

[7] I-129 (1.6×10^7 years) + n $\rightarrow$ I-130m (9 minutes) $\rightarrow$ I-130 (12 hours) $\rightarrow$ Xe-130 (stable) + e

[8] Cs-135 (2.3×10^6 years) + n $\rightarrow$ Cs-136m (19 seconds) $\rightarrow$ Cs-136 (13 days) $\rightarrow$ Ba-136m (0.3 seconds) + e $\rightarrow$ Ba-136 (stable)

[9] Cs-133 (stable) + n $\rightarrow$ Cs-134 (2.1 years) + n $\rightarrow$ Cs-135 (2.3×10^6 years)

[10] The reactions are: Pu-240 + n $\rightarrow$ Pu-241 (14 years); Pu-241 (14 years) + n $\rightarrow$ Pu-242 (380,000 years)

evident that neutron capture and the creation of heavier plutonium isotopes creates new problems in place of old ones. By contrast, when plutonium-239 fissions, most fission products are short-lived, while some are long-lived. Hence, significant reduction of the mass of long-lived actinides, such as plutonium, generally necessitates fission of the nuclei.

Fission transmutation reactions produce mostly short-lived fission products that decay into stable elements. The example below shows the production of two short-lived fission products, tellurium and molybdenum. They both undergo a series of beta decays. The decay chain of molybdenum-102 consists of short-lived radionuclides until it reaches stable (non-radioactive) ruthenium-102. Tellurium decays into long-lived cesium-135.

Pu-239 + n → Pu-240 → Te-135 (19 seconds) + Mo-102 (11 minutes) + 3 n

↓ ↓

I-135 (6.6 hours) + e Tc-102m (4.4 minutes) + e

↓ ↓

$Xe\text{-}135^{m}$ (15 minutes) + e Tc-102 (5.3 seconds)

↓ ↓

Xe-135 (9.1 hours) Ru-102 (stable) + e

↓

Cs-135m (53 minutes) + e

↓

Cs-135 (2.3×10^{6} years)

Proposed transmutation schemes

Various schemes have been proposed for transmutation. Three types of reactors (light water reactors, fast reactors, and sub-critical reactors) and two types of reprocessing have been proposed. Table 1 shows the type or types of reprocessing associated with each type of reactor and the radionuclides that would be candidates for transmutation. Most transmutation schemes would use a combination of reactors and associated reprocessing technologies. For example, in one scheme, light water reactors would be fueled with mixed oxide (MOX) fuel - that is, fuel made with plutonium extracted from conventional reactor spent fuel which is mixed with depleted uranium, with both materials being in an oxide chemical form. The MOX spent fuel then would be reprocessed and the transuranic actinides would be extracted to fuel a fast neutron reactor (also commonly called a breeder reactor). The fast reactor fuel would, in turn, be reprocessed and the remaining actinides would fuel a sub-critical accelerator driven reactor.

Table 1: Transmutation schemes

Reactors and neutron sources	Type of reprocessing and candidate radionuclides for transmutation	Comments
Light water reactors (LWRs) (the most common type of commercial nuclear reactor) The reactor is critical and fueled with either low-enriched uranium or mixed oxide uranium-plutonium fuel.	Reprocessing: aqueous Radionuclides: Primarily plutonium, Tc-99, I-129.	• Creates high proportion of higher mass actinides with associated severe radiation hazards • Reprocessing creates large amounts of liquid radioactive waste • Issues of reactor safety • Cannot fission most actinides • Heavy transuranic build-up, creating waste management problems
Fast reactors: The reactor is critical and can be fueled with plutonium, uranium or, potentially, fuel containing some minor actinides.	Reprocessing: mostly dry in advanced schemes. Radionuclides: Plutonium and possibly minor actinides. Tc-99 and I-129 may be possible but only in moderated targets outside the reactor core.	• The development of fast reactors has been crippled by persistent problems • Fission products are not efficiently transmuted • Heavy transuranic build-up though to a lesser extent than with LWRs • Issues of reactor safety
Sub-critical reactors: an accelerator-target system provides fast neutrons to a sub-critical reactor	Reprocessing: the reprocessing can be all aqueous or all dry or a combination of the two Radionuclides: plutonium and minor actinides. Tc-99 and I-129 may be possible but only in moderated targets outside the reactor core.	• Sub-critical reactors are only at the R&D stage • Cost is projected to be high. • Reactor safety still an issue • Fission products are not efficiently transmuted

None of these schemes can transmute uranium, cesium-135, carbon-14, and some other radionuclides. Table 2 (below) shows the various radionuclides of concern from the point of view of long-term management and their status with respect to various transmutation schemes.

Residual Waste

Even the most elaborate transmutation schemes will leave behind substantial amounts of long-lived radionuclides requiring disposal, while generating large new volumes of operating and decommissioning wastes. Transmutation does not eliminate the need for a high-level waste repository. First, no transmutation scheme is able to deal with all of the radionuclides of concern since many cannot be transmuted for practical purposes (see

example of uranium and Cs-135, above). Second, transmutation of Tc-99 and I-129 is not 100% effective, even with multiple passes through the reactor. Third, new long-lived fission products are created from the fission of the actinides. Fourth, fissioning of the actinides is not 100% effective in eliminating them. For instance, even the most optimistic, best-case estimate concedes that at least 2.4 metric tons of transuranic radionuclides would be left over after the transmutation of 906 metric tons of transuranics anticipated to be produced by US nuclear reactors during their licensed lifetimes.[11] Moreover, the composition of the residual transuranic waste would be shifted towards higher isotope actinides, making the residual fraction more radioactive per unit weight. This would result in greater radiological risks, complicate disposal, and limit any gains in repository capacity due to a smaller actinide inventory. Fifth, the disposal in a repository of cesium-137, which is mixed with cesium-135 in spent fuel, would necessitate a large repository. This is because the intense radioactivity of cesium-137 results in the generation of a large amount of heat, which necessitates an increase in spacing of the disposal canister. The large space requirements would negate one of the most important benefits of transmutation – that of reducing repository size for a given nuclear energy generation.[12] Only storage of long-lived wastes for a hundred years or more, with its attendant high uncertainties, risks, and costs, would significantly alleviate this repository capacity problem.[13] Finally, waste from prior reprocessing operations, whether for commercial or military purposes, is highly unlikely to be transmuted since almost all of it will have been vitrified for safety reasons before a transmutation program can be put into place. This large amount of waste would have to be sent directly to the repository. In other words, there are fundamental and substantial limitations to the reduction in long-lived radioactivity that can be achieved even with an elaborate and very expensive transmutation program.

Table 2 shows the main long-lived radionuclides of concern and the feasibility of their transmutation. As can be seen from this table there are a large number of radionuclides, which cannot be transmuted due to complicating factors or because of the nature of the radionuclide. These include the medium-lived fission products, uranium (which forms about 95 percent of spent fuel), and many long-lived radionuclides that arise either from fission or from neutron activation.[14] Of the long-lived fission products, only technetium-99 and iodine-129 have the potential to be fabricated into targets and transmuted in a reactor. The plutonium, and in some cases, the other minor actinides, would be made into fuel to run the transmutation reactor. The actinides could either undergo fission or

[11] ATW Roadmap 1999d. p. 38

[12] In this case strontium-90 would also likely be disposed of in the repository, since its half-life is about the same as cesium-137.

[13] For the first one hundred years the fission products dominate the radioactivity of spent fuel (with Cs-137 and Sr-90 being the predominant radionuclides). After 300 years it is the actinides which dominate the radioactivity. Both fission products and actinides contribute to the radioactivity in the period between 100 and 300 years (see NAS-NRC 1983, p. 30).

[14] Neutron activation refers to a process by which materials that are not originally radioactive become radioactive after being irradiated with neutrons (e.g. structural materials in the core of a reactor or the material that surrounds the reactor fuel).

Table 2: Main Long-lived Radionuclides of Concern

Radionuclide (half-life in years, to two significant digits)	Type	Impact	Transmutation Potential	Transmutation Problems
Tin-126 (100,000)	Long-Lived Fission Product	Groundwater release	Difficult	Difficult to separate from spent fuel/HLW. Long time to transmute. Lower isotopes result in new production of radionuclide
Selenium-79 (60,000)	Same	Same	None	Same
Cesium-135 (2.3 million)	Same	Same	None	Formation of more Cs-135 from Cs-133. Isotopic separation difficult due to presence of Cs-137
Zirconium-93 (1.5 million)	Activation Product	Groundwater release	None	Presence of stable Zr isotopes would produce more Zr-93. Would require expensive isotopic separation.
Carbon-14 (5,700)	Activation Product	Groundwater release and/or air release as CO_2; incorporation into living matter	None	Small neutron capture cross-section. Often released as gas from reprocessing operations
Chlorine-36 (300,000)	Activation Product	Groundwater	None	Presence of natural Cl-35 would generate more Cl-36
Technetium-99 (210,000)	Long-Lived Fission Product	Groundwater Release. Affects thyroid	Yes. Requires slow neutrons	Would require several transmutation cycles
Iodine-129 (16 million)	Long-Lived Fission Product	Same	Yes. Requires slow neutrons	Same. Also, difficulty in capturing during separation. Difficulty in fabricating targets. Could pose corrosion problems
Uranium (mainly U-238, 4.5 billion)	Actinide source material	Forms bulk of spent fuel (~94 percent by weight). Has higher radioactivity than TRU waste slated for geologic disposal	None. Would be separated and disposed of as LLW or used like depleted uranium	U-238 transmutation would result in the generation of more Pu-239 defeating the purpose of transmutation as a waste management strategy. Would essentially create a breeder reactor economy.
Americium-241 (430)	Actinide	Gamma-emitter. Human intrusion. Groundwater release (parent of U-233). Radiotoxicity	Preferably in fast reactors	Would require multiple separation and irradiation cycles. Would result in creation of curium which would make subsequent cycles more difficult
Neptunium-237 (2.1 million)	Actinide	Groundwater release	Preferably in fast reactor	Formation of more radioactive shorter-lived Pu-238
Curium-244(18)	Actinide	Highly radioactive alpha and gamma emitter. Contributes to heat of spent fuel.	Difficult. Requires fast reactor	Difficult to separate from other actinides in HLW due to handling and chemistry problems. Would require multi-recycling along with other actinides. Could require storage of decades or even a century. More Cm-244 and other Cm isotopes created in irradiation of lower actinides (Pu and Am).
Plutonium (mainly Pu-239, 24,000)	Actinide	Pu-239 Fissile. Radiotoxicity. Goes to bones	Fast reactor required for non-fissile isotopes.	Neutron capture forms higher isotopes and higher actinides (e.g. Am and Cm).
Strontium-90 (29)	Medium-lived Fission Product	Contributes to initial heat of waste. Determines repository capacity. Intrusion scenario dose. Behaves like calcium in the body	None	Cannot be transmuted due to small neutron cross-section. Forms a large part of the heat of spent fuel and high level waste and therefore limits increase in repository capacity from transmutation.
Cesium-137 (30)	Same	Same except behaves like potassium in the body. Also radiation barrier to proliferation.	None	Same. Also, separation from fissile materials eliminates radiation shielding for proliferation prevention.

Table is adapted and expanded from OECD/NEA 1999a, p. 470, and OECD/NEA 1999b.

capture a neutron, though for the purposes of transmutation, which is trying to reduce the amount of actinides, fission is preferred.

Transmutation would also create significant quantities of additional transuranic and low-level waste, particularly if aqueous reprocessing is used. Furthermore, it has been proposed in the United States to dispose of uranium separated from spent fuel in a transmutation program as "low-level" waste in shallow land burial sites. This, along with the possible shallow-land disposal of other long-lived radionuclides, could result in an even greater overall radiological risk to the public from transmutation, compared to disposal of all spent fuel in an appropriately selected and engineered repository. The same observation is also likely to be true of worker and public health hazards arising from repeated reprocessing of spent fuel, fabrication of increasingly radioactive fuels and operation of new reactor types with which there is little commercial experience. Transmutation, even in the context of a phase-out of nuclear power, would also require decades to implement and possibly centuries to complete.[15] This may require institutional control over the waste for time periods much longer than is feasible or desirable.

Implications of Transmutation

The implementation of any of the transmutation schemes discussed above would also have a number of implications for nuclear proliferation, the environment and human health, safety, cost, and the future of nuclear power.

Proliferation. All transmutation schemes require reprocessing and separation of transuranic radionuclides. The current use of commercial reprocessing and MOX fuel, the simplest of schemes to transmute a small fraction of existing plutonium, results in the separation of significant quantities of plutonium, which is undesirable from a proliferation standpoint. The current mismatch between reprocessing capacity and reactor capacity for MOX use has meant that a significant stockpile of commercial separated plutonium has accumulated worldwide (including 30 metric tons in Russia). While some new transmutation schemes would materials that would be unattractive to weapons designers in nuclear weapons states, they are nonetheless weapons-usable and would pose significant proliferation risks. Non-state groups or non-weapons states that do not have weapons-usable materials today might seek to acquire and use them because they may be more available in less secure facilities. Even the reprocessing methods that are labeled as proliferation resistant, such as pyroprocessing, can be modified to allow for the extraction of plutonium pure enough to make weapons.

Some reprocessing technologies proposed for transmutation may increase proliferation risks due to their compact size and attendant difficulty of detection. These would lead to new and more difficult problems in developing adequate safeguards in an already complex field. Furthermore, promotion of transmutation as a waste management tool may result in the widespread transfer of reprocessing technology. The separation of isotopes like neptunium-237 and americium-241 (which are two of the radionuclides produced during irradiation of fuel in a reactor) would also increase proliferation risks, since both of these radionuclides can also be used to make nuclear weapons. In sum, transmutation is a scheme that would greatly increase separation of weapons-usable material and/or the

[15] NAS-NRC 1996, p. 5 and OECD-NEA 1999b, p. 204. Some transmutation schemes would store medium-lived fission products for up to 600 years in order to allow them to decay (see Rubbia et al. 1997).

diffusion of technologies that would facilitate such separation. It will thereby considerably increase the risks of nuclear proliferation.

Environment and Health. Reprocessing, which is required in all transmutation schemes, is one the most damaging components of the fuel cycle. It results in the discharge of large volumes of waste and radioactive emissions to air and water. Health and environmental concerns regarding reprocessing are the basis of the demands of Ireland, Norway, Iceland, and Denmark that Britain and France eliminate their so-called "low-level" radioactive waste discharges from their reprocessing plants into the seas. The increased radiological risk of handling fuel that has been repeatedly irradiated is cause for serious concern. Finally, the increased transportation of high level waste required under a number of transmutation schemes would increase the probability of a transportation accident.

Reactor Safety. All transmutation schemes that would transmute significant amounts of plutonium and other transuranic materials require the use of reactors that are currently not commercial. Some schemes would use breeder reactors, which face serious technical issues even after five decades of development, and have not yet been commercialized. Other schemes would use accelerator-driven sub-critical reactors, which have not yet been built. Yet other schemes would use combinations of these two reactor types.

Some new reactors, notably accelerator-driven sub-critical reactors, have been described as "inherently safe." However, increases in certain safety features, in comparison with commercial light water reactors, is countered by decreases in other safety features and the creation of new safety problems particular to the new reactor designs. According to Dr. Lawrence Lidsky of MIT's Nuclear Engineering Department, "sub-critical systems can actually be more dangerous than conventional reactors if, as is often the case, there are more subsystems that can fail or initiate failures, and fewer backups. Probabilistic risk analysis is a complex art, requiring a deep understanding of possible accident initiators and accident progression, and the ATW design is far too rudimentary at this time to apply this powerful tool. However, it is clear that the currently envisaged ATW systems are more complex than fission reactors, have more accident initiators, and many fewer backup safety systems." It is thus premature, at best, to label these reactors as inherently safe. And according to one eminent authority, they could be a lot more dangerous. There is therefore ample reason for caution.

Cost. The cost of transmutation, particularly for the advanced schemes that would be required in order to have significant reduction of actinides, is prohibitively expensive (even in comparison to the billions to be spent on repository programs). Furthermore, while electricity would be produced to offset these costs, it is highly unlikely that these revenues will be sufficient. Transmutation would likely require tens of billions of dollars to develop, and additional large subsidies during operations, even after accounting for electric power sales. Even current uses of plutonium in reactors, both in light water reactors and in fast reactors, are not economical. The overall cost can be expected to be many tens of billions of dollars of net costs and overall investments up to hundreds of billions of dollars.

Continuation of Nuclear Power. Transmutation is not only considered in the context of managing the waste from the current generation of nuclear reactors (i.e. as part of a phase-out of nuclear power). Most transmutation schemes, particularly in Europe and

Japan, assume an indefinite continuation of nuclear power, with transmutation as one part of a new nuclear fuel cycle. By supposedly solving some of the current problems with nuclear power (particularly waste management, but also reactor safety in some cases), transmutation is seen by some as essential to ensuring the continued growth of nuclear power. Seen in this light, transmutation of waste is actually a Trojan horse for perpetuating nuclear power and hence the generation of more and more radioactive wastes for the indefinite future. This is surely not the way to solve the problem of managing radioactive waste from the current generation of commercial reactors.

Conclusions and Recommendations

Our main finding is that transmutation schemes will not solve long-term waste management problems. Well over 90 percent of the weight of spent fuel consists of uranium. According to according to current US proposals, the uranium would be treated as low-level radioactive waste and be disposed of in ways that will likely pose far greater risks than disposal in a carefully selected and engineered deep geologic repository. In addition, considerable quantities of transuranic materials would remain after transmutation, along with long-lived fission products. Large quantities of new waste would be created, along with new proliferation risks and high costs. Despite these severe limitations, transmutation continues to be seen by some as a “seductive” area of research and essential for revitalizing the “nuclear option.”

In light of these conclusions, IEER's main recommendation is that, because there is no sound technical basis for proceeding, transmutation should be abandoned as a waste management technology. Detailed findings and recommendations are given below.

Findings

1. Transmutation will not solve either the problem of long-term radioactive waste disposal nor the proliferation risks posed by current stockpiles of plutonium. While solutions are required for both of these problems, the use of reprocessing and nuclear reactors is not the best option.
2. The transmutation literature does not evaluate overall risk and is unclear about environmental or proliferation consequences relative to the once-through fuel cycle. The lack of comprehensive and consistent criteria by which to judge transmutation has led to a number of erroneous conclusions concerning its benefits.
3. Reprocessing is required for all transmutation schemes. Reprocessing is one of the most environmentally damaging parts of the nuclear fuel cycle, resulting in emissions to the air and water and in large volumes of radioactive waste. The increased separation requirements of transmutation means that even more processing is required as additional process steps are added to remove specific radionuclides.
4. The separation of radionuclides necessary for transmutation will increase proliferation risks by providing easier access to fissile materials. All separation processes, including those labeled "proliferation resistant," result in an increased proliferation risk over the once through fuel cycle. The implementation of transmutation as a waste management technology will result in more widespread application of reprocessing.
5. Transmutation can only be used to reduce the inventory of some of the radionuclides of concern for waste management. Even for those radionuclides, the process is not 100% efficient and significant amounts of long-lived waste will remain. Transmutation will not eliminate the need for a high-level waste repository or other form of isolation from the biosphere. The remaining long-lived radionuclides, including the uranium which accounts for about 94% of the spent fuel mass, as well as the radionuclides produced during the transmutation process will require disposal. Furthermore, transmutation can only be applied to spent nuclear fuel and some high level waste and not to the full range of radioactive wastes (e.g. transuranic wastes or mining wastes) which exist.
6. While the radiological risk from disposing of radioactive waste in a geologic repository may decrease as a result of transmutation, the overall risk to workers and the public may increase from a combination of disposal of separated uranium and other materials, emissions from new reprocessing and irradiation facilities, and processing of fuel that is more radioactive. These risks have not been adequately assessed in proposals for transmutation.
7. Transmutation will increase the mass and volume of radioactive material requiring disposal. In addition to the high level waste and uranium that would still require repository disposal (see Finding 5, above) reprocessing and transmutation operations will result in more transuranic and low level waste requiring disposal. These newly generated wastes will be in addition to the original mass of the spent fuel, resulting in

an overall increase in mass of waste to be disposed of.[16] Decommissioning wastes will also increase and can be expected to be substantial.

8. Transmutation will be expensive to implement. Life-cycle cost estimates are rarely presented, but current cost estimates which have been done are unrealistically low, particularly for reprocessing and decommissioning. Even with these low cost estimates and sales of electricity to offset those costs, full-scale transmutation will require some form of government funding and subsidy or substantial increase in utility waste disposal fees. In the United States alone, the net costs over the course of 118 years, after electricity sales, could be over $150 billion (as opposed to $36 billion for direct disposal at Yucca Mountain).[17]
9. Transmutation will rely on nuclear reactors that would pose serious hazards in case of accident. Both sub-critical and critical reactors contain large inventories of radioactive materials, which can be released during an accident. Transmutation, if it is to achieve any significant reduction in the inventory of actinides, will require the construction and operation of a significant number of fast reactors, whether critical or sub-critical, posing significant safety issues.
10. The increased radiological risks of working with reprocessed materials, particularly fuel that is repeatedly reprocessed, will increase risks to nuclear fuel cycle workers and increase the cost of protecting those workers.
11. Transmutation would require a sustained effort over very long periods of time. Assuming an immediate start to research and development activities, transmutation of the expected spent fuel from existing U.S. reactors would take 118 years to transmute (including development time). The Nuclear Energy Agency of the Organization for Economic Co-operation and Development estimates that transmutation could take decades, and even centuries, depending on various factors.
12. The reliance of some transmutation proposals on above-ground monitored storage for highly radioactive fission products for hundreds of years (e.g. in Carlo Rubbia's proposal for Spanish waste management) is unrealistic and risky.
13. Transmutation will increase the number of shipments of nuclear high level waste and therefore the probability of a transportation accident. Spent fuel or high level waste would have to be shipped from current storage locations to transmutation sites and then to final disposal. In cases where reprocessing facilities would not be co-located with reactors, the waste would have to be repeatedly shipped between reactors and reprocessing facilities. If transmutation does not begin until after a repository is opened and has started to accept waste (as would be the case in the United States), then spent fuel would be shipped from current storage locations to the repository,

[16] Though not addressed extensively in this report, it must be noted that each of the new facilities operated for the purposes of transmutation will eventually have to undergo decontamination and decommissioning procedures. This will result in even greater amounts of radioactive waste for disposal, including major components of the facilities such as the reactor cores. It is not clear how the increased radioactivity of fuel which has been repeatedly irradiated will affect the D&D process and the disposal requirements.

[17] ATW Roadmap cost estimate (ATW Roadmap 1999g) adjusted to reflect more realistic reprocessing costs as established by the National Research Council (NAS-NRC 1996). Figures are in undiscounted 1999 dollars.

removed from the repository for shipment to the transmutation site, and then the residual spent fuel and high level waste would be shipped back to the repository.

14. Transmutation of nuclear waste appears to be one component of a nuclear industry effort to increase the use of nuclear power. Significant development of nuclear power reactors would be required to implement transmutation and, at the same time, transmutation would be seen as a "solution" to the nuclear waste problem. The result could be a continuation of nuclear power, even beyond what would be necessary to transmute current reactor fuel, and thus a continual production of new nuclear waste. Hence, instead of reducing nuclear waste, it could result in increasing and continual generation of waste into the far future.

Recommendations

1. **Regulations governing the disposal of uranium should be strengthened.**
   The uranium extracted during transmutation has a higher enrichment than natural uranium and will be contaminated with fission products and actinides. The uranium will exceed the radioactivity concentration limit placed on plutonium waste in the United States many times over.[18] Despite this fact, transmutation proposals call for the uranium to either be used for commercial (e.g. as airplane ballast) or military purposes (e.g. armor-piercing rounds) or disposed of as low-level waste. None of these options would be protective of public health. Therefore, uranium should be regulated using the same criteria that are used for transuranic waste.

2. **The current use of plutonium fuel in nuclear reactors should be halted.**
   Transmutation schemes build upon the current use of plutonium in light water reactors as MOX fuel and on breeder reactor demonstration programs, which were supposed to produce more plutonium than they consumed. MOX fuel is uneconomical in comparison to other energy sources, such as wind power, and the use of MOX was only initiated when breeder reactor programs did not live up to expectations. Commercial MOX fuel use also increases proliferation risks due to the need for reprocessing in order to separate plutonium and complicates safety and environmental problems connected to reactor operation and waste disposal. Breeder reactor programs, which form the basis of a number of transmutation technologies, have been plagued by problems throughout their history, including safety deficiencies, technical operating problems, and uneconomical operation. They would pose even greater proliferation problems than the use of MOX in light water reactors, particularly as full-scale breeder reactor programs would result in even greater quantities of separated plutonium. Breeder reactors can also be relatively easily reconfigured from a waste transmutation role to one of making weapon-grade plutonium.

3. **Current reprocessing operations in all countries should be halted and commercial stockpiles of separated plutonium should be considered a waste to be immobilized.**
   Plutonium reprocessing operations pose unacceptable environmental, proliferation and financial risks and should cease. Existing stocks of separated plutonium should be immobilized (encasing it in a solid material like glass). This would reduce the proliferation risks of separated plutonium while not encouraging the further separation of plutonium from spent fuel. Feasibility studies should be conducted in the United Kingdom, France, and Japan (with the aid of the United States and Russia) on the conversion of MOX fuel fabrication facilities to ceramic immobilization facilities.[19]

[18] See Chapter V
[19] The issue of separated commercial plutonium will be further explored in a forthcoming report by IEER.

4. **The definition of reprocessing should be clarified**
   Any technology which processes spent fuel, and results in a product that includes separated fissile materials, or from which it is easier to separate fissile materials, should be considered a reprocessing technology. This is because virtually any combination of plutonium isotopes, as well as actinides such as americium and neptunium, can be used to make nuclear bombs. Thus, proliferation impacts should be evaluated according to the separation of weapons-usable materials and the potential of the technologies that are used for being modified for producing such materials even if that is not their normal function as part of a waste transmutation system.

5. **Waste management research efforts should be redirected towards scientifically sound long-term management of nuclear waste.**
   High-level waste management has been plagued by short-sighted political expediency. For instance, in the United States only one site, Yucca Mountain, is being actively developed, which has resulted in severe pressures to open it despite extensive evidence of its unsuitability. Reforms should be implemented to stop politically expedient repository projects, and those, like transmutation, which seem to have keeping nuclear power alive as a subterranean goal. We need a broad-based scientific search for appropriate disposal options in contrast to efforts on transmutation.

6. **Evaluations of transmutation should be based on the overall risks of such a program.**
   Much of the current technical literature on transmutation focuses on the possibility of transmutation to reduce the amount of actinides in high-level waste. This is a questionable approach, given the potential for significant increases in worker and public doses due to increased fuel cycle activities, inappropriate disposal of some reprocessing waste such as uranium, generation of more waste especially in reprocessing operations, and the open questions about the effect that transmutation will have on doses from a repository. All of these various risks need to be included in any overall analysis. At the very least transmutation programs should be suspended until such an analysis, conducted by an appropriate *independent* body, has been openly and thoroughly done with public input.

7. **Government funding of transmutation research should be stopped.**
   In Europe and Japan, where transmutation research budgets are substantial, funds should be redirected to repository programs or other nuclear waste management programs that do not rely on reprocessing and nuclear reactors. Transmutation programs are diverting valuable resources from other, more appropriate, waste management options. Similarly, in the United States, further work on Accelerator Transmutation of Waste (ATW) or other transmutation schemes should be halted. Furthermore, the United States Department of Energy should halt all research on separation processes, including those based on electrometallurgical techniques. This research should be considered a violation of the federal policy against reprocessing of commercial fuel.

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Chapter I: High-level Waste and Transmutation Schemes

For over fifty years, the nuclear industry has continually tried to advance itself as the ultimate energy provider. It was claimed that nuclear energy would be "too cheap to meter." As has been adequately demonstrated in a number of analyses (and in the electric bills of consumers of nuclear power) this claim never had a substantial technical foundation.[20] At the same time came the promise of "breeder reactors" which would eventually produce more plutonium than they consumed (breeder reactors are discussed extensively below). After spending tens of billions of dollars, most breeder reactor programs have been cancelled or are in serious jeopardy due to concerns over cost, non-proliferation, and poor performance.[21]

During the same period of time, waste from military and civilian nuclear reactors has been steadily accumulating with no definitive solution found so far. These wastes consist of both irradiated spent nuclear fuel from reactors and the high-level waste from reprocessing operations to remove plutonium for both civilian and military purposes.[22]

Spent fuel is a mixture of uranium, plutonium, other transuranic radionuclides and fission products. The uranium forms the bulk of the spent fuel mass and volume and is extremely long-lived. The plutonium, other transuranics, and fission products can either be long, medium, or short-lived depending on the radioisotope.

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[20] Makhijani and Saleska 1999

[21] See Makhijani 2000

[22] In the United States, and elsewhere, both spent fuel and the highly radioactive liquid waste generated by reprocessing spent fuel to extract plutonium, are classified as "high level waste." In addition, the solid waste created when the liquid high level waste is made in glass blocks (vitrified) is also called high level waste. Both the spent fuel and the solidified reprocessing waste are slated for geologic disposal. Therefore, depending upon the context, high level waste can refer to all of these wastes or it can refer to only the high level liquid waste (HLLW) or solidified liquid waste from reprocessing.

No country has yet opened a geologic repository for high-level waste, the spent fuel management option that is being pursued most vigorously in the United States and elsewhere.[23] International repository programs are at various stages of development.[24] Some countries have chosen initial repository locations and are in the process of evaluating and licensing those repositories. Yucca Mountain, in the United States, is one such example. Other programs are using underground laboratories to evaluate different geologic media in order to make a decision. Some countries are still at the stage of simply developing criteria for underground disposal and are relying solely on aboveground laboratory programs. In some cases, such as the United Kingdom, an explicit decision has been made not to begin the siting process for many decades.

Geologic repositories are a very difficult enterprise. Since the radionuclides of concern have half-lives of thousands and even millions of years, the performance of the repository must be estimated over extremely long time-frames. The planning for repositories must also account for the possibility of accidental or intentional human intrusion. As a result of the difficulties of repositories and problems with the management of the process of finding suitable repository sites, there has been significant opposition to repositories.

The lack of progress in managing the increasing volume of high-level nuclear waste has been one of the factors involved in the decline of nuclear power in the United States.[25] Transmutation proponents hope that a solution to the waste problem will remove a major barrier to new nuclear power plants (this is discussed further in Chapter V). In addition, it is perceived that the new reactors that would be required to dispose of current stocks of waste will form the basis for new generations of nuclear reactors based on new designs and concepts.

Interest in the possibility of transmuting nuclear waste has grown in the last decade, particularly as some of the more optimistic proponents claim it will eliminate the need for repositories (this claim is discussed further in Chapter V). The concept is to use one or more of several types of nuclear reactors to convert long-lived radionuclides into short-lived radionuclides. This makes waste management more predictable. There is also some value to bringing the risks of waste management closer to the generations that created them. However, in order to transmute, it is necessary to separate the radionuclides of concern in high-level waste. This problem, among others, greatly complicates the prospects for transmutation.

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[23] The United States government has opened a repository, the Waste Isolation Pilot Plant, for transuranic (TRU) waste arising from nuclear weapons production and other activities in the Department of Energy complex. Transuranic waste is defined by the Energy Department as waste containing over 100 nanocuries per gram of radionuclides with atomic numbers greater than 92 (uranium) and whose half-lives are greater than twenty years. In general this mainly consists of plutonium waste, but can also include significant quantities of americium and curium. Unlike high level waste, which can either be spent fuel or reprocessing waste consisting mainly of fission products, this waste is generally produced during operations which involve handling plutonium, and other actinides, such as occur in the production of nuclear weapons.

[24] "International Repository Programs," S*cience for Democratic Action* Vol. 7, No. 3, May 1999. p. 14-15.

[25] Other factors specific to nuclear power include safety and proliferation concerns. Additionally, economic considerations have played a large role.

In the meantime, the volumes of radioactive waste continue to increase as reactors discharge spent fuel and reprocessing operations continue in countries such as the UK and France. Commercial reactors have discharged approximately 220,000 metric tons of spent fuel since the beginning of commercial nuclear power.[26] There are approximately 400 nuclear power reactors currently operating worldwide. This adds about 10,000 metric tons of highly radioactive spent fuel per year to the total.[27] In the United States alone, past reactor operations have only discharged about half of the total expected under current operating licenses.[28]

Nuclear Fuel Cycle

In order to fully understand the role that transmutation proponents see it playing in the future of the nuclear industry, it is necessary to have some knowledge of the nuclear fuel cycle. Generally speaking there are two different types of nuclear fuel cycles: Open and Closed. Open fuel cycles use nuclear fuel once in reactors and then the spent fuel is considered waste. This fuel cycle is also referred to as the "Once-Through Fuel Cycle" and these terms are used interchangeably in this report. Closed fuel cycles attempt to make use of the fissile material which remains in the spent fuel.

In both cases, the fuel cycle begins with the mining and milling of natural uranium. The milling process is responsible for 95% of the volume of radioactive waste from the nuclear fuel cycle and contains a number of long-lived radioisotopes.[29] The natural uranium consists of 99.284% U-238 and 0.711% U-235 along with trace amounts of U-234. U-235 belongs to a group of radionuclides called "fissile," which means that they can fission even with neutrons of low energy and thus can sustain a chain reaction more readily (see Appendix A). The majority of nuclear reactors operate with low-enriched uranium (LEU), meaning that the natural uranium has been processed to increase the percentage of U-235 up to 3-5%.[30] This also creates a waste by-product left over from the enrichment process, which is known as depleted uranium (because it has even less U-235 than natural uranium).

In a moderated nuclear reactor such as the Light Water Reactor (LWR) in common usage worldwide, the graphite reactors of the former Soviet Union, and the heavy water reactors developed by Canada, the U-238 which forms the bulk of the fuel mass is not fissioned (U-238 will undergo fission in fast reactors without a moderator

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[26] As of 1995 approximately 180,000 t of spent fuel had been discharged worldwide (IAEA 1997b, p. 119). Between 1995 and 2000 a further 10,000 metric tons per year has been discharged for a total of approximately 220,000 t.

[27] Energy Information Administration, U.S. Department of Energy. *World Annual Spent Fuel Projections by Region and Country, Reference Case, 1998-2020*. http://www.eia.doe.gov/cneaf/nuclear/n_pwr_fc/data98/spentfuel.html.

[28] As of December 1998, U.S. commercial reactors have discharged approximately 38,500 metric tons of spent fuel (DOE 1998, p. 4). If all currently operating reactors conclude their current operating licenses a total of 87,000 metric tons of spent fuel will have been discharged by 2035.

[29] DOE 1997, p. 0-14 (and Chapters 5 and 6). This includes both commercial mill tailings and waste classified as 11e2 by-product material, which consists of waste from uranium processing.

[30] Reactors can also use natural uranium (e.g. CANDU reactors) or high-enriched uranium (above 20% U-235 for research and test reactors and 97.3% for U.S. naval reactors). HEU is also weapons-usable (enriched to about 93.5% U-235). See Bodansky 1996, p. 88 and Cochran et al. 1987, p. 125

however). While U-238 cannot be fissioned as easily as U-235 in these reactors, it does absorb neutrons to produce plutonium-239. Pu-239, a fissile isotope like U-235, also undergoes fission as it builds up in the reactor and accounts for a significant portion of the fission energy (particularly in reactors operating on non-enriched natural uranium such as the CANDUs). After the fuel is removed from the reactor, there remains both some U-235 and Pu-239. In countries with open fuel cycles, the spent fuel remains at the reactor site waiting for shipment to a repository.

The closed fuel cycle attempts to make use of the remaining Pu-239 by extracting it through a complicated and environmentally hazardous process called reprocessing. The reprocessed plutonium can then be used either for nuclear weapons use or for use in nuclear reactors.[31] The plutonium can be put back into the same type of reactor in the form of a Pu oxide – U oxide (also called MOX or Mixed OXide fuel). However, the use of MOX in light water reactors should not really be considered a closed fuel cycle. In fact, it can be more accurately characterized as a twice through fuel cycle since the plutonium can only be passed back through the reactor a limited number of times (due to the change in isotopic composition as more neutrons are absorbed to produce heavier elements). In practice, this has limited MOX to only one more pass through the reactor.

The other possibility is to fuel a reactor designed to eventually produce more plutonium than they consume (these reactors are called breeder reactors). A fuel cycle using such reactors could be closed, but after years of research and tens billions of dollars these programs have largely been abandoned, scaled back or put on hold. For more information on these fuel cycles and the different types of reactors refer to Appendix C.

Thus, both open and closed fuel cycles produce high-level waste. In the open cycle, this is simply the spent fuel from the reactor. In the case of MOX cycles, the high level waste (HLW) consists of both reprocessing waste and spent fuel. For a closed cycle based on fast reactors, the high level waste would be from reprocessing operations.

What is Transmutation?

Transmutation is the transformation of one isotope into another (or two others). It involves a change in the number of nucleons (protons and/or neutrons in the nucleus of the atom). Transmutation, in the context of nuclear waste management, generally involves the absorption of a neutron to either create the next heaviest isotope of an element or to fission the target element into two or more fission products. Transmutation can transform a stable element into an unstable, radioactive element, and vice versa and it can transform a long-lived radionuclide into a short-lived one, and vice versa.

To understand transmutation one has to know what the building blocks of atoms are, and what role they play in determining the characteristics of a particular element.

[31] While production of plutonium for nuclear weapons is done in such a way as to minimize the amount of higher plutonium isotopes, and therefore has a different isotopic composition than reactor-grade plutonium, it is generally accepted that reactor-grade plutonium can be used to manufacture a nuclear weapon. While such a weapon would not be of the same sophistication as a weapon designed with weapons grade plutonium, it would still provide a sizable explosion (see Chapter VI for more details). See NAS 1994 and NAS 1995 for more details.

The atoms of all elements are made up of neutrons and protons.[32] The number of protons is what distinguishes one element from another. For example: carbon has six protons and nitrogen has seven. A given element can have atoms with different number of neutrons; in that case the element has isotopes. For example: chlorine has two stable isotopes, chlorine-35 (17 protons and 18 neutrons) and chlorine-37 (17 protons and 20 neutrons). However, chlorine-36 (17 protons and 19 neutrons) and chlorine-38 ((17 protons and 21 neutrons) are both radioactive. All the isotopes of a given element have the same chemical properties which make it very hard to separate them from each other. However in some elements, the nuclear properties, such as how long it takes for the isotope to decay, differ between isotopes. For example: in spent fuel there are five isotopes of cesium. One is stable and the half lives for the other four vary from short to long.

Each isotope has a characteristic decay time called the half-life. The term half-life is used because it measures how long it takes for half of the atoms to decay. For example, the half-life of plutonium-239 is 24,000 years and that is the time it will take half of the Pu-239 to decay into U-235. After another 24,000 years half of the remaining Pu-239 will have decayed into U-235 (leaving one quarter of the original amount of Pu-239) and so on. Thus, while it is never possible to tell when a particular nucleus of Pu-239 will undergo decay, it is possible to know how long it will take a group of Pu-239 atoms to decay.

Transmutation occurs:

- in nature. An example is the spontaneous decay of uranium-238 into daughter products,
- in artificial devices, such as reactors. Two examples: (i) the splitting of one atom of uranium-235 in two lighter fission products; (ii) the absorption of neutron by uranium-238, followed by two decay steps results in the creation of plutonium-239.

In the context of waste management transmutation refers to a small subset of such nuclear transformations: those that convert long-lived radionuclides into short-lived radionuclides or into a stable element. However, the same process that transmutes long-lived radionuclides into short-lived radionuclides can also induce nuclear reactions to convert short-lived radionuclides to long-lived ones. This complicates the prospects for transmutation as its purpose is to shorten the long-term risks of spent fuel management

The case of cesium is a good illustration. As noted above, there are 5 isotopes of cesium in spent fuel, and since chemical separation of these isotopes from each other is virtually impossible, transmutation involves all of them. The isotope of greatest concern from the point of view of longevity is cesium-135, whose half-life is 2.3 million years. It can be transmuted (through neutron absorption) into a short-lived radionuclide (Cs-136) which, in turn, decays into a stable radionuclide (barium-136). However during that process, some cesium-133, a stable isotope gets transmuted into cesium-134 and then cesium-135.

[32] An exception is hydrogen, the simplest element, which has only one proton.

For transmutation to proceed in a nuclear reactor the long lived radionuclides have to be separated from the fission products and to various degree from each other, this, for several reasons:

- the fission products would prevent the chain reaction from occurring,
- as noted above, some short lived fission products would be transformed into long lived radionuclides, and this must be prevented
- depending on the subsequent transmutation method chosen, the long-lived radionuclides will have to be separated from each other to various degrees, so that the specific transmutation characteristics of each can be taken advantage of.

Because it is possible to transmute radionuclides of concern (either through neutron absorption or fission), proponents of transmutation present it as an answer to the problem of long-term management of spent fuel. This report will survey the various technologies and briefly analyze their implications from the point of view of cost, safety, waste management aspects, and proliferation.

Composition of high level waste

In order to understand how proposed transmutation claim to solve the nuclear waste problem it is necessary to provide a generic overview of the composition of spent nuclear fuel and the radionuclides of concern in spent fuel and reprocessing waste. This will be followed by a general overview of transmutation as a solution and the technologies involved.

The definition of transmutation in the Introduction applies to the transformation of only one element into another element (or two elements) and is unidirectional: from long lived to short-lived. However, spent fuel is a mixture of many different radionuclides with half lives ranging from less than a second to half lives comparable to the age of the earth. When spent fuel from a typical light water reactor is unloaded from a nuclear power plant, its composition by weight is typically[33]:

- 95.9 % uranium (of which approximately 0.8% is U-235)
- 3.2 % fission products
- 0.7 % fissile plutonium
- 0.2 % of non-fissile plutonium

There are also smaller amounts of heavy radioactive elements called minor actinides.[34] At the time of unloading the spent fuel is very radioactive and would give a lethal dose to a person standing close to it. Short-lived fission products contribute the

[33] See Lamarsh 1983, Figure 4.25, p. 150. This is for fuel initially enriched to 3% U-235 and for a 1,000 MWe plant operated at 75% capacity. Changes in any of these parameters can change the figures given.
[34] The actinides are a group of elements on the periodic table which include, most importantly, neptunium, plutonium, uranium, americium, and curium, among others. However, often uranium and plutonium are considered separately because of their importance. Therefore, the rest are sometimes classified as the minor actinides. Another term, transuranic, is used in U.S. waste regulations to specify certain actinides heavier than uranium and present in certain concentrations.

most to the radioactivity, which decreases rapidly as the short-lived fission products decay. One hundred and fifty days after unloading, the contribution of the fission products to the radioactivity of the spent fuel has decreased by a factor of 30, and after ten years by a factor of 400.[35] For the first 70-100 years after unloading the radioactivity is dominated by the fission products. Both fission products and actinides contribute to the radioactivity over the next 100 to 300 years. Thereafter the actinides are the main contributors.[36]

In addition to spent fuel, highly radioactive waste from past and on-going reprocessing operations also need to be accounted for. These reprocessing operations were undertaken to remove plutonium for use in either nuclear weapons or in reactors, such as MOX fueled light water reactors. Countries which have undertaken major reprocessing, for either military or commercial purposes, and which will have to account for these wastes in implementing transmutation include the United States, Russia, France, and the UK. In addition, Japan, which has actively pursued transmutation research, has had a modest reprocessing operation, is constructing a larger reprocessing facility, and currently has its spent fuel reprocessed in the UK and France (for which it must take back the waste). The radioactive inventory of reprocessing waste differs from that of spent fuel. Most obviously, it does not contain significant amounts of plutonium or uranium as they have already been separated. In addition, some of the fission products are volatilized during reprocessing (e.g. Iodine-129) and are, at least in part, emitted into the air or water from the reprocessing facility. Furthermore, the physical composition of the waste is different from spent fuel. Some is still in liquid form, stored in large tanks, while some has been vitrified into glass logs. Thus, reprocessing waste poses its own unique challenges for transmutation, including for the separation of radionuclides of concern.

The potential difference this could make to the feasibility and efficacy of any transmutation proposal being implemented in a particular country can be more readily observed by comparing the waste type and amounts in the United States and France. Table 3 provides a rough comparison for both spent nuclear fuel and liquid high level waste. The figures provided here are not exact but are meant to convey the differences in between the two countries as a result of commercial and military reprocessing in France as compared to the United States. While U.S. reactors and reprocessing facilities were clearly separated between commercial and military, such a distinction was not always made for France. Therefore, both commercial and military spent fuel and reprocessing waste inventory are included in the "commercial" category for France. This appears to be justified as only two metric tons of plutonium, out of a total of 84 tons of plutonium extracted, have been used for the weapons program.[37]

To put these figures in context, it should be noted that there are currently a little over 100 reactors operating in the United States and 58 reactors operating in France. In the United States only a small fraction of the spent fuel from commercial reactors has been reprocessed while France has reprocessed approximately 17,000 metric tons of the 30,000 metric tons of spent fuel discharged. As can be seen from the table, the amount of

[35] Benedict, Pigford, and Levi 1981, Table 8.1, pp. 354-356
[36] NAS-NRC 1996, p. 323 and NAS-NRC 1983, pp. 29-30
[37] WISE-Paris 2000, p. 10.

spent fuel in the United States is roughly three times that in France. However, the volume of commercial reprocessing waste in France is significantly higher than in the United States. Some of this waste has already been vitrified into solid glass. The economics of pursuing further separation and transmutation activities on either the liquid or solidified high level waste would have to be questionable considering the small amount of fissile fuel contained therein to provide electricity for sale.

Table 3: Comparison of U.S. and French Spent Fuel and High Level Waste Inventories

Country	SNF Mass (MTIHM)		HLW Volume (m^3)	
	Military	Commercial	Military	Commercial
United States[a]	2,483	34,252	345,300	2,000
France[b]		13,000		85000

MTIHM-Metric Tons of Initial Heavy Metal

[a] Military inventories are from DOE 1997, p. 1-11 (spent fuel, June 1997) and p. 2-12 (high level waste, end of fiscal year 1996). Commercial inventories for the United States are from DOE 1997, pp. 1-7 (commercial spent fuel) and 2-23 (commercial HLW at the West Valley Demonstration Plant) and are for the end of calendar year 1996.

[b] Spent nuclear fuel mass from WISE-Paris 2000, p. 1 of English version of Conclusions, as of December 1998. High level waste volume has been calculated by us using a nominal 5 cubic meters of high level waste per metric ton of spent fuel processed (as per NAS-NRC 1983, p. 34). This is the volume prior to vitrification.

Characteristics of the radionuclides considered for transmutation

The radionuclides considered for transmutation are various isotopes of plutonium, neptunium, americium and curium for the actinides, and iodine-129 and technetium-99 for the fission products (as discussed elsewhere, the other long-lived radionuclides such as Cesium-135, Chlorine-36, and Selenium-76 are not considered for transmutation). However, curium, which is hard to separate from americium, is a problem radionuclide because it is a source of neutrons and a strong gamma emitter.[38] Curium interferes with fuel and target fabrication, and with the recycling of americium targets.[39] Curium-242, with a half-life of 163 days, has virtually disappeared after 3 years and is only a potential problem in transmutation systems with short cooling times (e.g. accelerator-based systems).

Since the half-lives of curium-242, curium-243, and curium-244 are "relatively" short, there have been a variety of proposals as to how to handle the curium problem by allowing some to decay away. One is to store spent MOX fuel for approximately fifty

[38] Gamma radiation is electromagnetic radiation (like ordinary light or X-rays), but with high energy (X-rays have energies equivalent to the lowest gamma ray energies). Gamma radiation is highly penetrating and causes damage to biological organisms by ionizing atoms. A gamma emitter is a radioisotope that results in the emission of gamma radiation when it decays. Protection from gamma radiation is necessary in some operations in the nuclear industry (for example by using remote handling).

[39] In some case the americium would be part of the fuel and mixed homogeneously with the plutonium, but in other cases, the americium would be separated and formed into special targets for irradiation (similar to the way the fission products, technetium and iodine, would not be part of the fuel but would be made into targets).

years to allow some of the curium to decay into plutonium before further processing for transmutation.[40] However, it is not clear how this would alleviate the problem of curium production during transmutation. If fifty years of cooling is required after every transmutation cycle (or even after just the first two or three transmutation cycles), the timeframe for transmuting the waste is well over a century. Other proposals would separate the curium from the americium after processing MOX fuel and then store the curium for approximately a century.[41] In addition to creating storage problems over long periods of time, this would not reduce the problem of separating curium. If the curium and americium are instead stored together, a material will have to be developed that can both withstand the high radiation of the curium during storage, but can also then act as a fuel element (so as to avoid another processing step before irradiating the americium in a reactor). Another option is to reduce curium production by have very long-burn-up times and high neutron fluxes. This illustrates some of the difficulties with transmutation, even for those elements (i.e. the actinides) that transmutation is supposed to be able to handle the best.[42]

The characteristics of the radionuclides considered for transmutation, as well as few important ones that cannot be transmuted are shown in Table 4. A discussion of the various headings and the data are below.

Half-lives and amounts

Plutonium is the most abundant radionuclide to be produced in irradiated fuel, with Pu-239 being the dominant radionuclide in terms of the number of grams produced per ton of fuel.[43] The plutonium isotopes have a range of half-lives, as do the other actinides that are produced. The actinides contribute to both medium term heat of the spent fuel, as well as to the long-term radioactivity of the spent fuel.

The long-lived radionuclides most often considered for transmutation, Tc-99, I-129, and Cs-135 are shown Table 4. As can be seen, their half-lives are in the hundreds of thousands and millions of years. Other isotopes of Cesium are also shown to illustrate why Cs-135 transmutation is not feasible. Cs-133, which is stable, can absorb neutrons and become Cs-135. Cs-137 is a strong gamma emitter, which precludes, mainly for health and safety reasons, practical isotopic separation of the three cesium isotopes and thus makes transmutation of Cs-135 impossible.[44] Sr-90 is also shown because of its important contribution to the short and medium term radioactivity from the spent fuel.

[40] OECD-NEA 1999b p. 49
[41] OECD-NEA 1999b, p. 38
[42] Salvatores and Zaetta 1997, pp. 111-112.
[43] Note that this is for radionuclides *produced* during irradiation. Uranium is still the dominant radionuclide, comprising around 96% of the spent fuel mass.
[44] The strong gamma emissions from the Cs-137, aside from whatever effect it may have on the separation process itself, poses a risk for workers. As a result, the shielding and safety measures that would be necessary if cesium isotopic separation were to be attempted would make the process extremely expensive.

Table 4: Characteristics of Radionuclides in Spent Light Water Reactor Fuel Considered for Transmutation or Long-Term Monitored Storage

Isotope	Content (g/t)	%	half-life years	Decay	Cross Section					
					Thermal			Fast		
					σ_f	σ_c	σ_c/σ_f	σ_f	σ_c	σ_c/σ_f
Pu-238	600	1.6	86	α	2.4	27.7	12	1.1	0.58	0.53
Pu-239	22,300	58.7	24,400	α	102	58.7	0.58	1.86	0.56	0.3
Pu-240	8600	22.6	6,580	α	0.5	110.6	221	0.36	0.57	1.58
Pu-241	4600	12.1	14.4	β	94.8	36.7	0.38	2.49	0.47	0.19
Pu-242	1900	5.0	3.79×10^5	α	0.43	29	67	0.23	0.44	1.9
Np-237	430	100	2.1×10^6	α,γ	0.52	33	63	0.32	1.7	5.3
Am-241	220	67	430	α,n	1.1	110	100	0.27	2.0	7.4
Am-242	0.7		16 hours	β	159	301	1.9	3.2	0.6	0.19
$Am\text{-}242^m$			141	β	595	137	0.23	3.3	0.6	0.18
Am-243	100	31	7400	α,γ,n	0.44	49	111	0.21	1.8	8.57
Cm-242			0.446		1.14	4.5	3.9	0.58	1.0	1.7
Cm-243	0.3	1	28	α,γ,n	88	14	0.16	7.2	1.0	0.14
Cm-244	21.4	94	18	α,γ,n	1.0	16	16	0.42	0.6	1.4
Cm-245	1.2	5	8500	α	116	17	0.15	5.1	0.9	0.18
Cm-246	0.2		5500	α,n						
Tc-99	953		2.12×10^5	β		13.8				
I-129	247		1.7×10^7	β		3.2				
Sr-90			28.1	β		1.34				
Cs-133			Stable			158				
Cs-134			2.05	β		129				
Cs-135			3×10^6	β		30.2				
Cs-137			30.0	β,γ		0.176				

Isotopes: m=metastable, %=isotopic percentage of particular element (e.g. Pu-239 accounts for 58.7% of the plutonium)

Decay: α=alpha; γ=gamma; β=beta; n = emits neutrons due to spontaneous fission.

Cross Sections:σ_f = cross section for fission; σ_c= cross-section for neutron capture

Source: Bataille and Galley 1998, Tableau 29 for neptunium, americium and curium (present in uranium oxide fuel with a burn up of 33 000 MWd/t, three years after unloading) and Tableau 2 and 3 for plutonium content. Cross-sections for the actinides are from OECD-NEA 1999b, Table II.3, p. 148. Thermal refers to a typical thermal reactor rather than thermal neutrons at room temperature. NAS-NRC 1996 (pp. 24 and 50) was used for Technetium-99, Iodine-129, Strontium-90, and Cesium-137 cross-sections. The cross-section for Tc and I is for the actual spectrum of neutrons in a pressurized water reactor (e.g. an LWR). The cross-section for Sr and Cs is for room temperature thermal neutrons (and would likely be lower for the slightly higher energy neutrons in a thermal reactor or moderated target in a fast reactor). OECD-NEA 1999b, p. 47 was used for the content of Tc-99 and I-129 in spent fuel. Benedict, Pigford, and Levi 1981, p. 361 was used for Cs-133, Cs-134, Cs-135 data (cross-section data is for the typical neutron spectrum in a PWR).

Cross sections

The cross section for neutron absorption of a particular radionuclide measures the probability with which it will be transmuted by absorbing a neutron.[45] The bigger the

[45] The unit for the cross section is the barn, which is equivalent to an area of 10^{-24} m^2. The physical cross section of a nucleus is a circle with the radius of that nucleus. However the nuclear reaction cross section of a nucleus, which is the probability with which a neutron will collide with that nucleus, has little to do with the size of the nucleus. Rather, it is a function of the structure of the nucleus as well as the energy of

cross section the greater the chance a particular radionuclide will have to get transmuted. In the case of fission or activation products, such as technetium-99 or zirconium-93, the neutron capture cross section is the probability that it will absorb a neutron to create the next heaviest isotope (e.g. technetium-100). For the actinides, there are two cross sections which compete at the same time, one for neutron absorption followed immediately by fission (called the fission cross-section), the other for neutron capture resulting in the creation of a radionuclide of higher mass (the neutron capture cross-section). The size of the cross sections are themselves a function of various factors, including the structure of the nucleus (how many protons and neutrons it contains) and the speed at which a neutron strikes a particular radionuclide. For waste management purposes transmutation by fission is the outcome that is desired, or stated another way a high ratio of fission over capture. This is because it takes a number of radioactive decays for any actinide to reach a stable isotope. If the actinide captures a neutron it will be no closer to stability than it was before. However, if it is fissioned, the fission products, which are also radioactive, decay into a stable isotope in less steps. For example, the capture of a neutron by U-235 can result either in fission or in the formation of U-236, which then stays in the spent fuel as a contaminant. The U-236 decays (with a half-life of 2.34×10^7 years) into thorium-232.[46]

For all the actinides the ratio of fission over capture is greater in a fast reactor than in a thermal reactor. Therefore fast reactors are more efficient transmuters than thermal reactors for the actinides.[47]

As can be seen, transmutation of the fission products by neutron capture is difficult due to the small cross-sections. There is essentially no absorption of neutrons by fission products when the neutrons are of high energy ("fast") and the cross-sections are not listed. For thermal neutrons, the cross-sections are small but are not zero. For the medium-lived fission products such as Sr-90 and Cs-137 the cross-sections are too small to consider transmutation by neutron capture. The long-lived fission products, such as technetium-99 and iodine-129, do have slightly larger cross-sections than the medium-lived fission products. As this would require thermal neutrons, light water reactors would be the reactors of choice. However, with a fast reactor it may be possible to moderate and slow the neutrons down just outside of the core and place targets of technetium and iodine there (these are called ex-core targets).

Decay modes

Appendix A provides an overview of the differing types of radioactive decay. With the exception of plutonium-241 and Am-242, all the actinides listed in the above table are alpha emitters whereas the fission products are beta emitters. Some of the

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the neutron that strikes the nucleus. For example, carbon-12, has a cross section of 0.0034 barns, whereas boron-10 has a cross section of 3837 barns for thermal neutrons. Although carbon-12 and boron-11 have about the same radius, one is a neutron moderator and the other a neutron absorber because of their nuclear structure.

[46] Thorium-232 has an even longer half-life (1.4×10^{10} years).

[47] Fast fission also produces more neutrons per fission, which helps counteract the lower fission cross-sections. Fast reactors also use a higher enrichment and sometimes denser fuel to increase the fission rate. Extra neutrons were also a factor in the development of breeder reactors based on fast reactors, since breeding requires neutrons for both breeding and to maintain the fission chain reaction.

actinides and fission products are also gamma emitters, neutron emitters or both. As will be discussed in the next section the introduction of gamma and neutron emitters in fuel fabrication poses serious health and safety problems.

Categories of Materials

There are, broadly speaking, three categories of materials of concern in spent nuclear fuel (the possibility of transmuting materials in each category is discussed further below in this chapter):

a) Medium-lived fission products: After the spent fuel has cooled in the reactor pools in order to allow some of the very short-lived radioactivity (less than five year half-lives) to decay away, there still remain some important fission products with intermediate half-lives. The two most important are strontium-90 (29.1 year half-life) and cesium-137 (30.17 year half-life). These two isotopes pose two particular problems. First, these isotopes contribute the most to the short-term radioactivity (for approximately the first 70-100 years) for scenarios of intrusion into the repository. Second, the heat due to their radioactive decay limits the loading of the repository per unit volume.[48] These radionuclides cannot be practically transmuted.

b) Long-lived Fission Products: A portion of the radioactivity in the spent fuel comes from isotopes with very long half-lives (up to millions of years). These isotopes contribute little to the radioactivity from the spent fuel in comparison to either the shorter-lived fission products or the actinides. However, long-lived fission products dominate the long-term dose in some repository scenarios due to their much greater solubility (which results in their transport into the human environment).[49] This category of materials includes Technetium-99 (210,000 year half-life) and Iodine-129 (16 million year half-life), which are proposed for transmutation, as well as other fission products such as selenium and tin, which are not viable candidates for transmutation.[50] Though not strictly fission products, we have also included some other long-lived radioisotopes present in spent fuel, such as chlorine-36 and carbon-14, in our discussions of long-lived radionuclides. Those long-lived radionuclides that are not proposed for transmutation have generally been excluded for practical reasons, rather than because they are not a risk. Those reasons include difficulty of separation from other radionuclides, difficulties in handling the material, small neutron cross-sections, the presence of a stable isotope which will transmute to create more of the radioactive isotope in question, and long transmutation half-lives[51]

c) Plutonium and Minor Actinides: Spent fuel also contains plutonium and other actinides (see table above). In addition to affecting the heat loading of the repository (especially after 100-300 years), the actinides have the highest

[48] NAS-NRC 1996, p. 23
[49] NAS-NRC 1996, p. 23
[50] NAS-NRC 1996, p. 23-24 and OECD-NEA 1999b, p. 48
[51] The amount of time it takes for half of the radionuclide to be transmuted. A small neutron cross section and the production of the radioisotope from fission in the reactor or by a lower isotope capturing neutrons are generally the reason for long transmutation half-lives.

radiotoxicity (as measured in terms of Sieverts/Becquerel or in the amount of air or water to dilute the radionuclide below the regulatory limit) of the long-lived radionuclides to be placed in a repository.[52] This is due to a variety of factors, including the amount of actinides produced (sometimes called the "inventory"), their specific activity, and their radiological effect once they have entered the body. However, in contrast to their high radiotoxicity, most repository scenarios show low doses for the actinides, particularly plutonium, due to high retardation factors (i.e. they are absorbed by the geologic media and do not reach the human environment). There are a number of questions about the validity of these scenarios however. For example, plutonium migration at the Nevada Test Site adjacent to Yucca Mountain has occurred much faster than had been believed possible.[53] A few actinides, under certain conditions, are already known to play a significant role in the expected dose from certain repositories (e.g. Np-237 in Yucca Mountain).[54]

Some of the actinides are also fissile materials that can sustain a nuclear chain reaction. These include U-235 from the original fabrication of the fuel (and from decay of Pu-239), as well as fissile isotopes, which build up in the fuel during irradiation. The most notable of these is Pu-239, but also includes americium-241 and neptunium-237. All can sustain a chain reaction and are weapons-usable in separated form. These materials pose a severe proliferation risk, particularly Pu-239, and must be closely safeguarded. The "elimination" of these actinides, particularly Pu-239, is considered to be an important goal of transmutation and seen by some as the only means to eliminate their proliferation risk. This is discussed further in Chapter V. Also, studies have been conducted assessing the criticality risk in a repository due to materials rearranging after time into a configuration that could result in a nuclear explosion.[55]

The radionuclides of concern and their contribution to the dose from a repository will vary depending on the geologic media and the type of repository. health must also be considered. shows the radiation dose as a function of the time it takes water to travel in a basalt repository. As can be seen from the figure, the dose in this particular case is dominated early on by Lead-210, then by Carbon-14, and then by Iodine-129. Other important radionuclides include Ra-226, Cs-135, Se-79, as well as three actinides, Np-237, Pu-239, and Am-243. This can be compared with Figure 3, which shows that the radiotoxicity of spent LWR fuel is dominated by the actinides. However, the actinides are not considered to be as soluble as the fission products and, thus, the dose from the repository is dominated by the fission products (with a substantial contribution from the

[52] Sieverts is a measure of radiation dose while the becquerel is a measure of radioactivity (and is equivalent to one disintegration per second). Thus, Sv/Bq is a measure of the radiation dose one receives from being exposed to a given amount of radiation. The number of Sv/Bq differs between the radionuclides as explained in the text.

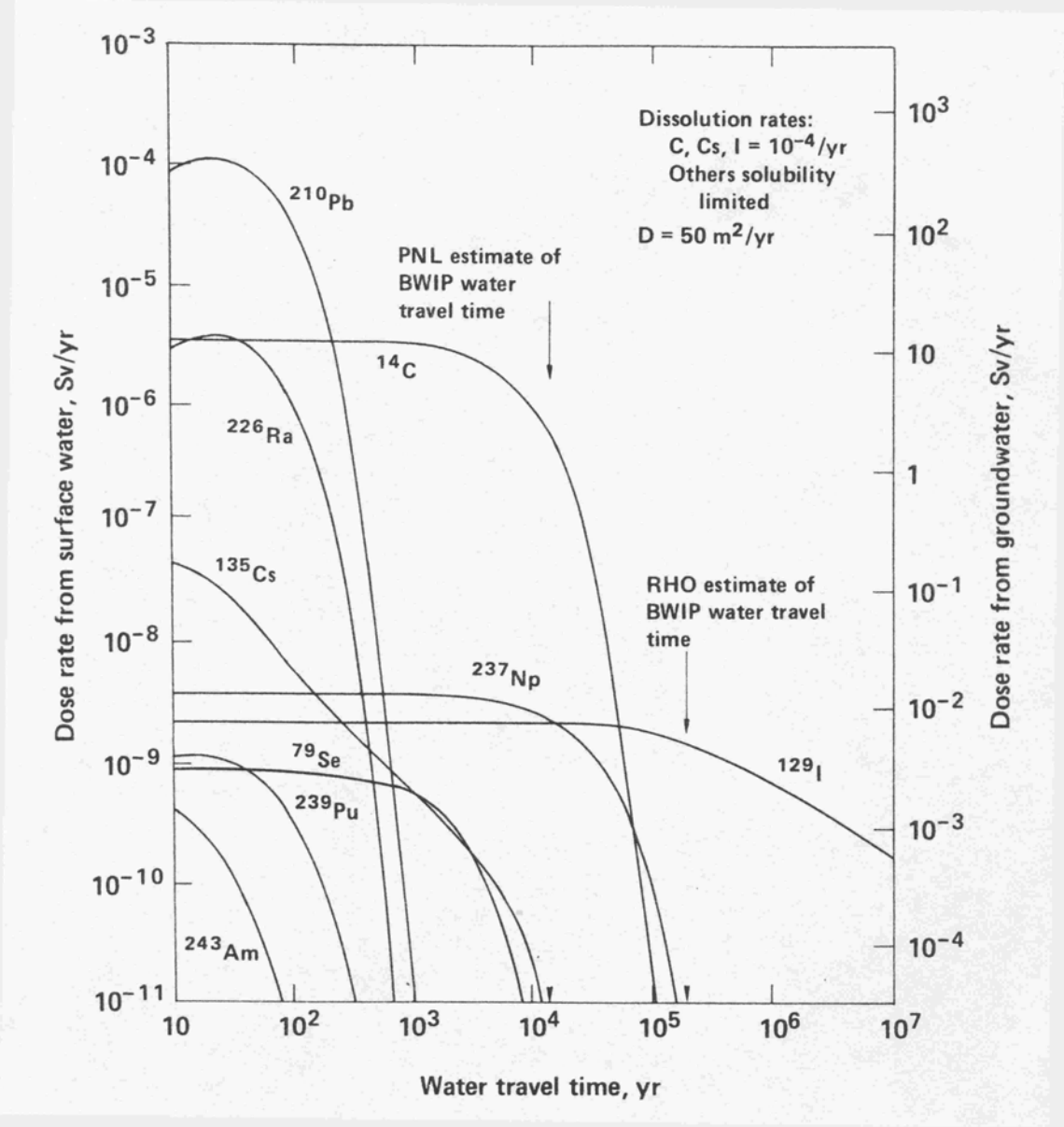
[53] Kersting et al. 1999. Also recent experiments have shown that, contrary to previous results, there is a soluble form of plutonium oxide which is formed at low temperatures (up to 350 ^{o}C). See Haschke, Allen, and Morales 2000 and Madic 2000. Accounting for this soluble PuO_{2+x} in which the plutonium is in a +VI state may change the repository dose scenarios.

[54] NAS-NRC 1996, pp. 329, 331, 335. In particular, the table on p. 331 shows that Np-237 could be the third or fourth largest contributor to the dose in an unsaturated tuff repository such as Yucca Mountain.

[55] See Bowman and Venneri 1994.

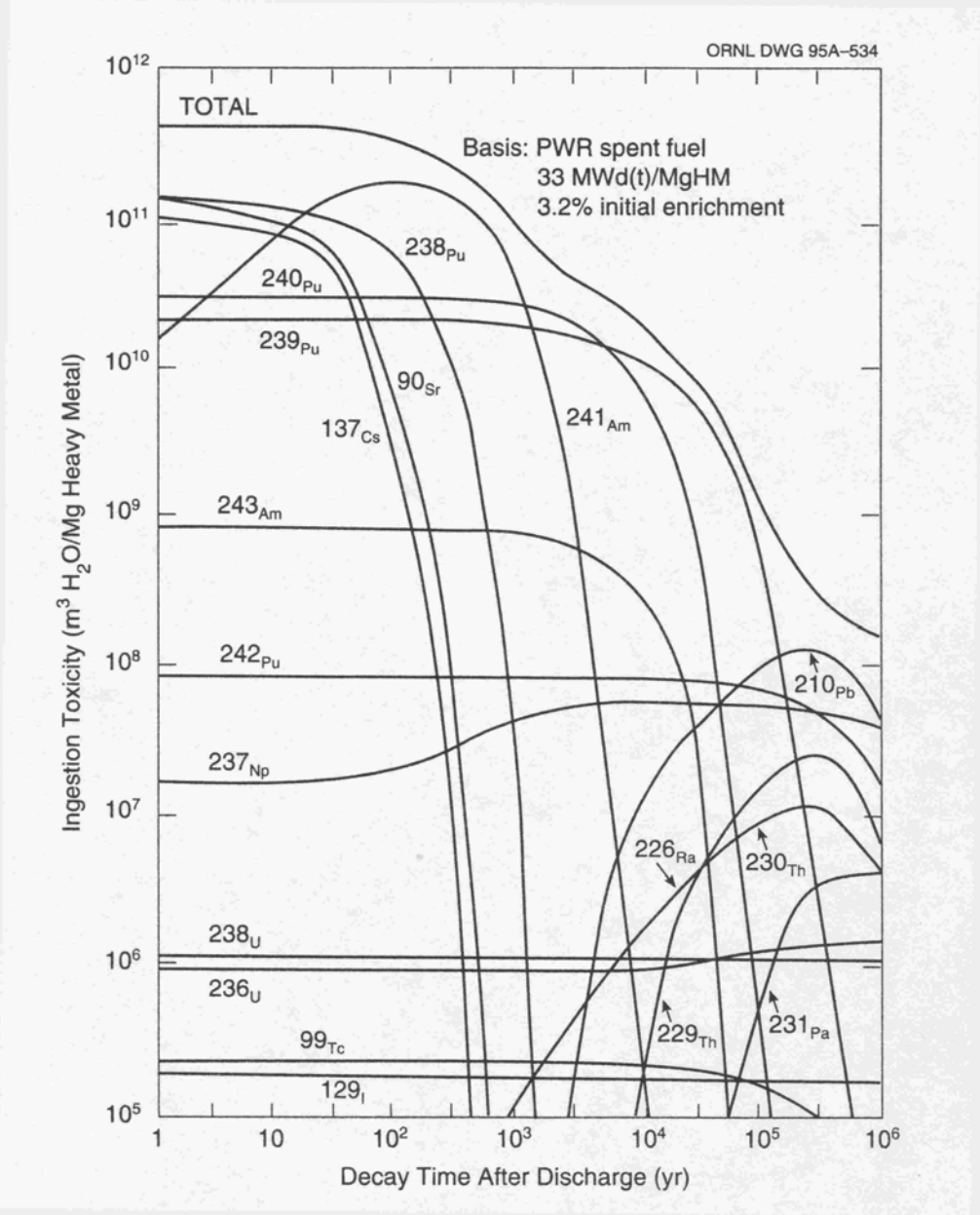
medium lived fission products, strontium-90 and cesium-137, early on). Thus, in determining the value of transmutation, it is not sufficient only to consider the radiotoxicity of the waste. The actual transport of the radionuclides through the environment and their potential effects on human health must also be considered.

Figure 2: Estimated Doses from a Repository in Basalt



Source: NAS-NRC 1983, Figure 9-14, p. 283

Figure 3: Radiotoxicity of Radionuclides in Spent LWR Fuel



Source: NAS-NRC 1996, Figure 2-2, p. 24

It should be reiterated that the dose from the repository will depend on the form of the waste placed in the repository, the contents of that waste, and the particular geology of the repository.[56]

[56] See Tables G-2 and G-3 of NAS-NRC 1996 (pp. 330-331) for a good illustration of this fact. Table G-2 presents relative doses due to different radionuclides in the case of a granite repository while G-3 provides the same for a tuff repository. Not only are the relative importance of the radionuclides different for the two geologic media, they are different even if the media is the same depending on the model used. Thus, a Finnish estimate for granite repositories places the order of importance as I-129, Pa-231, C-14 while the British estimate has I-129, Tc-99, and Cs-135 as the top three radionuclides.

Transmutation as proposed solution

One solution that has been proposed to deal with (primarily) the long-lived fission products and actinides has been termed Separations and Transmutation (S&T). Simply put, the spent fuel would undergo processing to separate it into different waste streams. Some long-lived fission products (LLFP) and some actinides would then be irradiated using a large neutron number of neutrons. The goal would be to fission the majority of the actinides and to transmute those long-lived fission products into short-lived radioisotopes by neutron absorption. Transmutation schemes provide separation of radionuclides and a neutron source in different ways, but these two underlying concepts are common to all schemes. The rest of the actinides and fission products, as well as any new waste created would have to be disposed of and some would go to a repository.

Transmutation Basics

There are four main components to any transmutation system. First, are separation facilities that process incoming spent fuel from conventional nuclear reactors or existing liquid high level reprocessing waste and process fuel from the transmuter. Second, is a source for the neutrons needed to transmute the long-lived fission products and fission the actinides. Third, is a reactor into which the long-lived fission products and actinides are placed. In some cases, such as light water reactors and breeder reactors, the neutron source and the transmutation reactor are the same. In other cases, such as accelerator-based schemes, the neutron source and reactor are linked but physically separate.[57] Fourth, there needs to be waste management facilities to handle both the separations waste, the radionuclides which cannot be transmuted, and the residual waste since the transmutation process is not 100% effective. There are broadly four types of transmutation methods that are being investigated:

- aqueous chemical separation followed by transmutation in light water reactors,
- aqueous chemical separation followed by transmutation in fast breeder reactors,
- pyroprocessing separation (dry process) followed by transmutation in fast breeder reactors,
- pyroprocessing separation (dry process) followed by transmutation in accelerator driven reactors.[58]

Fission and Transmutation of actinides

Actinides can undergo two basic reactions when subject to a large flux of neutrons: fission and neutron absorption into a higher atomic weight isotope. This is true

[57] Of course, even in these cases the fission of actinides in the reactor provides an additional source of neutrons.

[58] We are not considering a fifth type, phototransmutation, in this report. In phototransmutation an electron accelerator produces photons which photofission the actinides resulting in neutrons which can transmute the fission products. The photons can also knock out neutrons from the nucleus. Most of the basics are the same between phototransmutation and accelerator driven neutron transmutation, however, the research efforts in phototransmutation are significantly smaller. See Friedlander et al. 1981, pp. 157-158 for more information about photonuclear reactions in general.

for both fissile and non-fissile isotopes. However, in some cases an isotope is more likely to undergo one or the other. For example, Pu-239 is a fissile isotope and is more likely to undergo fission than neutron capture (to form Pu-240). However, neutron capture does occur and this is the source of build-up of higher actinides in ordinary reactor fuel. Higher actinides include high-energy gamma emitters. Additionally, the proportion of a particular radioisotope undergoing fission versus capture will change depending on the energy of the neutrons (called the neutron spectrum). Generally speaking, high-energy or "fast" neutrons have a higher probability of causing fission in the actinides. Lower energy or "thermal" neutrons will fission some of the actinides (e.g. Pu-241), but overall, the result is a greater shift of the inventory of actinides to higher isotopes and elements in comparison to the fast reactor.

The goal of transmutation with respect to the actinides is to fission the vast majority of them present in the original fuel while minimizing production of actinides from neutron capture. This establishes a number of parameters for performance. Uranium separation must be near perfect in order to prevent the production of new plutonium and higher actinides. If a thermal spectrum is being used to transmute fission products, then the requirements on separation of minor actinides is very high to prevent a shift to higher mass numbers.

However, even after transmutation there will still remain actinides that must be sent to a repository. For example, if the United States were to use the ATW system described in a recent Roadmap to transmute the 87,000 MT of commercial spent fuel expected to be produced by current reactors during their present license periods an estimated 2.4 tons or more of transuranics will remain in the end. This would be a decrease from the 900 or so tons originally in the spent fuel, but it would still be a significant amount..[59] This would still contain significant amounts of Pu-239. Furthermore, the isotopic mix of the transuranics would be shifted to higher actinides. The higher actinides, such as americium and curium, can have long- half-lives (in the hundreds of thousands of years), but are generally shorter-lived than plutonium or uranium. Thus, the higher actinides have more radioactive decays in a given period of time and are therefore more radioactive (and therefore hotter) than plutonium. Thus, the heat load and specific radioactivity of the final waste would be much higher (per unit mass). This can be considered to be a best case scenario since it utilizes accelerators to achieve high fission rates. Reactor based transmutation schemes would have a much higher final inventory of actinides since they rely on the actinides to maintain criticality and thus cannot achieve high burn-ups during the final phase. The length of time required to achieve a certain reduction in the amount of actinides is discussed further in Chapter V.

One actinide that poses a particular problem for transmutation is uranium and it is not included as one of the actinides when discussing actinides to be put in a reactor for transmutation. However, the vast majority of the mass of spent nuclear fuel is composed of uranium-238 (approximately 94 percent). Neutron irradiation of U-238 would simply result in the production of more Pu-239 and defeat the one major purpose of

[59] ATW Roadmap 1999d, p. 37-38. 2.4 tons will be sent to the high level waste repository from processing the fuel. In addition there will be a residual amount of transuranics left in the last reactor core. This is discussed further in Chapter V.

transmutation, which is to eliminate, to the greatest degree possible, the plutonium. In the United States, for example, current transmutation proposals would either dispose of the uranium as low-level waste or re-use the uranium. As is discussed further in Chapter V, neither of these options would be protective of public health and the environment. The uranium should be disposed of in a geologic repository. However, if uranium were disposed of in a geologic repository, it would negate some benefits of transmutation (such as a reduction in the mass of waste to be disposed of in a repository).

Transmutation of long-lived fission products

Long-lived fission products can undergo transmutation by absorbing a neutron. For example, the two long-lived fission products most often considered for transmutation undergo the following reactions (half-lives shown in parentheses):

Tc-99 + n → Tc-100 (16s) → Ru-100 + e

I-129 + n → I-130 (12.4h) → Xe-130 + e

Both Ru-100 and Xe-130 are stable. Equally as important, since they themselves could capture a neutron and form the next heaviest isotope, the next two higher isotopes of both are stable. Additionally, I-127, which is also present would undergo transmutation to I-128. Iodine-128 would then decay (with a 25 minute half-life) to Xe-128, which is also stable.[60] However, the difficulties in capturing the iodine that is released during reprocessing and anticipated problems in fabricating suitable targets from the captured iodine may result in a decision to release the iodine to the environment (air and/or water) during reprocessing operations. The current practice in France is to discharge iodine to the sea.[61]

The transmutation rate of the long-lived Tc-99 and I-129 will depend on a variety of factors, including the type of reactor used. For example, in thermal light water reactors, the transmutation rate is approximately 11% and 3% respectively per year.[62] This slow rate is due to the relatively small cross-section and the production of new Tc and I in the fuel. The Nuclear Energy Agency (NEA) estimates that it takes approximately 30 years to transmute half of the long-lived Tc-99 .[63] Putting Tc and I in a thermal reactor also requires changes to the reactor fueling in order to compensate for the loss in reactivity (e.g. by over-enriching the fuel). In a fast reactor (including fast neutron accelerator based reactors), the cross-section for absorption of Tc and I is even smaller (see Table 4) which leads to a need to have a moderated target area (e.g. surrounding the fast neutron core). Calculations based upon the flow of materials in the U.S. ATW roadmap indicates an average transmutation rate of approximately 1.5% per year for Tc-99.[64]

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[60] NAS-NRC 1996, p. 50.
[61] OECD-NEA 1999b, p. 36
[62] NAS-NRC 1996, p. 50
[63] OECD-NEA 1999b p. 52
[64] The 87000 MTHM of spent fuel is estimated to contain 73.1 MT of Tc-99. The final waste is estimated to contain 5.34 MT of Tc-99 for an overall transmutation of 92.7%. This is for 8 1/2 ATW stations operating for 60 years. See ATW Separations 1999d, pp. 37-38.

Another long-lived fission product candidate for transmutation is Cs-135. However, Cs-135 would pose a significant challenge for transmutation. While the Cs-135 would transmute to stable Ba-136, there would be production of Cs-135 by neutron capture on Cs-133, which has a higher neutron capture cross-section, resulting in a net *production* of Cs-135. Therefore, Cs-135 transmutation would require isotopic separation of the cesium. However, the Cs-137, which is also present, is an intense gamma emitter and would make that sort of processing very risky.[65]

It should also be noted that the benefits of transmuting some long-lived fission products, particularly Tc-99, is tied to design and choice of a repository. As with Np-237, Tc-99 would be much less soluble (and therefore less likely to enter into the groundwater) if a repository with reducing conditions were chosen.[66] This further demonstrates the need for more research into different repository conditions. The relationship between repository design choices and expected benefits of a transmutation program are discussed further in Chapter V.

Unlike technetium and iodine, the other important long-lived radioisotopes (e.g. Cs-135, Se-79, Sn-126, Cl-36, and C-14) are not being considered for transmutation. It is not feasible to transmute these radioisotopes despite their important contributions to the dose from certain repositories. Some, such as Sn-126, are accompanied by stable isotopes (Sn-116, Sn-118, Sn-119, Sn-120, Sn-122, Sn-123, and Sn-124). Neutron capture on these stable isotopes can lead to more Sn-126 production. This is also the case with Cs-135 as discussed above and Cl-36 (Cl-35 is stable). Others, such as C-14 have very small neutron capture cross-sections. For Se-79, one of the difficulties is separating it from the rest of the high-level waste due to its chemical behavior.[67]

There are four options being considered for non-transmutable radionuclides, depending on the isotope. Some gaseous isotopes like C-14 could end up being released to the air during reprocessing operations. The others, including gaseous radionuclides like Kr-85, could either be sent to the repository, stored aboveground, or disposed of as low-level waste. Aboveground storage is being proposed in some cases for both cesium and strontium. In the case of cesium this would have a dual purpose. This would allow the Cs-137 to decay and then isotopic separation of the Cs-133 and Cs-135 could be performed followed by transmutation of the Cs-135. It would also allow the Cs-137 to be disposed of as low-level waste. For strontium, low-level waste disposal would be the goal. However, this would require aboveground storage for hundreds of years. In some transmutation proposals, other long-lived radionuclides would also be stored above ground along with the cesium and strontium. Transmutation proponents are advocating

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[65] NAS-NRC 1996, p. 50.

[66] One characteristic of a repository is whether the conditions in the repository are "reducing" or "oxidizing." It is beyond the scope of this report to explain oxidation and reduction reactions, however, it is sufficient to know that these reactions (in which one reactant gives up electrons and one takes electrons) changes the chemical form of the elements involved. This affects the solubility of the radionuclide of interest and therefore its mobility. For example, in oxidizing waters technetium is mainly in the form TcO_4^- which is highly soluble. However, under reducing conditions, that chemical form of technetium would be reduced to TcO_2-$2H_2O$ (s), where (s) indicates a solid. In other words, the technetium changed chemical form and precipitated out of the water, meaning that it would not be as mobile in the environment. For more information about this subject see Langmuir 1997, Chapter 13 (example comes from p. 521).

[67] OECD-NEA 1999b, p. 48, Volckaert et al. 1999, p. 470, Bowman 1997, pp. 142-145.

the low-level waste disposal option for many radionuclides since it would reduce the load on the repository. This is an absolute criterion for those proposing that transmutation can eliminate the need for a repository. As will be discussed further in Chapter V, the use of current low-level waste burial practices to handle those radionuclides that cannot be transmuted is a major disadvantage of transmutation proposals. Furthermore, the separation and transmutation processes themselves will create more low-level waste to be disposed of.

Furthermore, it is not clear that those transmutation proponents who advocated LLW disposal have determined exactly how that is to be accomplished. For example, a 1997 International Atomic Energy Agency Status report on accelerator transmutation included a paper on meeting low level waste regulations.[68] According to the author, in order for most of the long-lived radionuclides to meet the Class C limit for low-level waste, anywhere from 90% to 99% of the radionuclides would have to be separated and transmuted. However, there was no discussion of how exactly that was to be accomplished, given the difficulties described above. Another approach, contained in the proposals for another accelerator based system, called the Energy Amplifier, is to dilute the waste until it meets the low-level waste limit (see Chapter IV and V).

Medium-lived fission products

While it is theoretically possible to transmute the medium-lived fission products, Sr-90 and Cs-137, it is not practical.[69] First, these radionuclides have very small neutron capture cross-sections (i.e. they do not readily absorb neutrons to transmute to a higher isotope). Second, as discussed above, putting Cs in the transmutation reactor would result in a net build-up of long-lived Cs-135. Therefore, most transmutation proposals would separate Sr and Cs before dealing with the other fission products and the actinides. The strontium and cesium would then be placed in specialized containers. Three options have been identified for the medium-lived fission products once they have been separated from the rest of the spent fuel. The first is storage aboveground in engineered facilities for up to 600 years until the radioactivity of the Sr-90 and Cs-137 have decayed to the levels set by LLW waste disposal regulations. The second is to send the containers to a long-term repository along with the high level waste. The third is to send them to a separate medium term repository designed specifically for storing such wastes.

Aboveground storage: One of the major benefits cited for aboveground storage is that it reduces the impact of early intrusion scenarios and allows for more waste to be placed in the repository due to much lower heat levels.[70] A further benefit cited is that, since the Cs-137 radiation level would be low, long-lived Cs-135 could be separated from the stable Cs-133 and then put through a transmutation reactor.[71]

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[68] Bowman 1997, pp. 140-145

[69] NAS-NRC 1996, p. 23 and OECD-NEA 1999b, p. 216

[70] See for example, NAS-NRC 1996, p. 326

[71] This scenario is not considered by either the National Academy Panel (NAS-NRC 1996) or in the recent OECD/NEA status report (OECD/NEA 1999b) which assume disposal of the Cs-135 after the decay of the Cs-137. However, some transmutation proponents have suggested the possibility that after the decay of the cs-137, the Cs-133 and Cs-135 could be separated in order to transmute the Cs-135. See, for example,

There are also a number of disadvantages to aboveground storage. First, is its implications for the barrier against theft or diversion of plutonium for weapons purposes. The difficulty in obtaining plutonium is often described using the term "the spent fuel standard." This means the plutonium is just as difficult to retrieve as the plutonium in spent fuel. An important component of the spent fuel standard is the fact that spent fuel is highly radioactive, making it difficult to process, and difficult to steal. Cs-137, a high-energy gamma emitter, forms a significant portion of that radiation barrier. Removal of Cs-137 to storage would mean that the radiation barrier in a repository would be lower, increasing the risk of deliberate intrusion.

Second, there are a number of uncertainties to storing radioactive waste aboveground for such long times. This is discussed further in Chapter V. Third, their disposal as low-level waste is problematic, especially as it is likely to be done in conjunction with LLW disposal of long-lived fission radionuclides, and should not be considered a viable option. This is discussed further in Chapter V.

Long Term Repository: The medium lived fission products could also be sent to the repository for high level waste. This would not reduce the short-term heat load of the repository and thus could call into question one of the main stated advantages of transmutation, namely that a higher portion of waste could be placed in the repository and a second repository could be avoided.[72] It also means that the thermal stresses of a hot repository would remain, making prediction of the repository performance difficult (see Chapter V). The one positive effect that separation of the medium-lived fission products would have is that they could be placed in a solid matrix ideally suited for the individual element in order to minimize environmental risk.[73]

Medium Term Repository: In this scenario the strontium and cesium would be packaged and sent to a different repository than the high level waste, presumably one sited and constructed specifically to help contain these radionuclides. However, the repository would be designed with the purpose of containing the waste for only 500 years or so. While avoiding the most serious problems of aboveground storage, there would still be a number of potential problems that would have to be considered. First, a new repository would have to be sited, licensed and operated. Second, this does not take into account the long-lived Cs-135 and any other long-lived contaminants. Third, the problem of removing one of the theft barriers (Cs-137, discussed above) would exist. Fourth, it would change the basis for the current long-term repository (as discussed above).

Rubbia et al. 1997b, p. 63. While favoring the disposal of the Cs-135 as low-level waste after the Cs-137 has decayed, Rubbia et al. also note that future generations may wish to transmute the Cs-135 instead.

[72] For example, Laidler 1999 shows that the volume of ceramic waste containing medium-lived fission products would in fact be greater than the original volume of spent fuel. In addition, there would be depleted uranium oxide waste from the front-end process that would have to be managed. In effect, the waste volume would be *increased* rather than *decreased* by transmutation. Information published in the final Roadmap report for ATW indicates a small net decrease in the mass of waste to be sent to the repository if one does not include the uranium (see discussion of repository capacity in Chapter V).

[73] As noted in the OECD/NEA Status report, a number of fission products could be processed into particular chemical or metallurgical forms that would improve their performance, but management of these various separate forms of individual radionuclides could be more difficult than handling one high level waste form. OECD/NEA 1999b, p. 205.

This idea has also been discussed in the context of a program that would implement separations without transmutation. In other words, the differing radionuclides would be separated to the extent possible but without transmutation in order for each type of radionuclide to be packaged in a more suitable matrix. This would pose even greater risks and exacerbate all of the problems discussed above. In particular, this would imply that the transuranics would be separated and disposed of in the long-term repository without transmutation and without a significant component of their radiation barrier. The long-lived fission products would also be disposed of without transmutation calling into question the advantages of such a program for environmental protection or health reasons.[74] Given the amount of processing required and the attendant environmental, worker and public health risks from such processing it is questionable whether any overall advantage would be gained from repackaging the radionuclides.

Unwanted processes

In addition to all the desired transmutations that would occur, there would be a number of undesirable nuclear reactions, which would create new radionuclides of concern.

- As was discussed above, the successive neutron capture of Cs-133 to Cs-134 and then Cs-135 excludes Cs from being considered for transmutation. Similar problems occur with trying to transmute chlorine-36 or selenium-79.
- As with any nuclear reactor with a large number of neutrons, these neutrons will irradiate the reactor components themselves resulting in the reactor becoming radioactive. Decontamination and decommissioning of the large number of facilities proposed in some transmutation schemes would be a major undertaking.
- In the case of accelerator based systems, there would also be spallation products. These are the residual nuclei left after the neutrons have been "boiled off" in the spallation target. These are further discussed in Chapter V due to their potential health hazards and for their implications for waste management.

Separation

As is discussed in more detail in Chapter II, all transmutation schemes require both front-end chemical processing to separate the spent fuel into different streams and repeated reprocessing of the transmutation fuel. Some proposals take advantage of existing separations processes while others would require the large-scale development of processes currently only developed on the laboratory scale.

Separation necessary for Transmutation: In order to accomplish its goals transmutation will require very precise and very efficient processing with minimal losses and cross contamination in order to avoid unwanted processes and to send as little waste as possible to a repository. Uranium, mainly consisting of U-238 will have to be

[74] The argument in favor of this proposal is that individual radionuclides would be put into a physical form most suited for isolating it from the environment. However, if the waste is then placed in shallow land burial, it is not guaranteed that the improved waste form would compensate for the reduction in isolation from the environment.

efficiently separated (greater than 99%) in order not to produce more plutonium (which, of course, would defeat the purpose if the goal were to "eliminate" the plutonium). Similarly, plutonium and the minor actinides to be fissioned must be efficiently separated from the fission products so they can be transmuted. Inefficient separation would result in larger amounts being buried in a repository. In addition, there are certain undesirable radionuclides that must be removed. For example, cesium must be removed to avoid even larger build-up of long-lived Cs-135 from stable Cs-133.

i) Existing Processes: Currently, the most widespread separation technology is based upon the PUREX process. This is an aqueous process using large volumes of acids to chemically separate the plutonium and uranium from the fission products and minor actinides. Currently, neptunium, an important contributor to the dose from some repositories, can also be extracted by adjusting the chemical process.[75]

ii) New Processes: New processes for separations are being developed in order to meet the need to separate fission products and all of the actinides for transmutation. Different processes are being developed to separate long-lived fission products and minor actinides from liquid high level waste and spent fuel. Some of these are aqueous processes similar to PUREX (e.g. TRUEX to separate transuranic elements from HLW) while others use new non-aqueous processes (e.g. pyroprocessing, also known as electrometallurgical treatment) in an attempt to avoid some of the major problems with PUREX or to be able to use fuels for which PUREX is not well suited.

Consequences of Separation: As discussed in more detail in Chapters II and V, separation processes produce additional waste. In particular, PUREX results in large volumes of liquid waste contaminated with highly radioactive fission products. PUREX reprocessing to separate plutonium for weapons use at the Hanford and Savannah River Sites has created some of the most intractable of clean-up problems in the Department of Energy complex. PUREX also results in separated plutonium, which poses severe proliferation risks. Pyro-processing would entail smaller volumes, but this also means higher concentrations of radioactivity posing additional problems for worker safety. Also, while the proponents of pyro-processing point to the lack of separated plutonium, it does create separated transuranics, which can undergo further separation to isolate the plutonium. All separation processes have adverse proliferation consequences. This is discussed further in Chapter V.

Neutron Sources and Reactors for Transmutation

Nuclear reactors are generally classified according to the energy of the neutrons used. The neutrons produced during fission have a high energy and are called "fast" neutrons.[76] Neutrons can be slowed down by collision with a moderating material and

[75] Albright and O'Neill 1999, p. 89.

[76] The energy of fission neutrons can vary widely. For the fission of U-235 by slow neutrons (as in a current nuclear reactor), the neutron energy is usually between 1 and 2 MeV. Bodansky 1996, p. 64.

become "thermal" neutrons.[77] An ideal moderator is close in weight to a neutron and the most common have been water and graphite. The basic difference in fission with a fast versus thermal spectrum is in how easily the fissile material fissions, which isotopes will undergo fission, and how many neutrons are produced per fission. In general, fissile isotopes have a lower fission cross-section in a fast spectrum, but produce more neutrons per fission. For this reason, fast spectrum reactors have been favored for breeder reactor programs. In a thermal spectrum, fewer neutrons are produced per fission, but the fissile isotopes have a higher fission cross-section.[78] For a variety of reasons, the development of thermal spectrum reactors has been less difficult than fast spectrum reactors.

This report divides its discussion of neutron sources and reactors between critical reactors, which act as both neutron source and transmutation reactor and can sustain a chain reaction on their own, and sub-critical reactors, which use accelerated protons to generate supplemental neutrons through spallation to sustain the reaction and to transmute some radionuclides. Critical reactors in use today use a wide variety of technologies in terms of the types of fuels and coolants used as well as their neutron spectra. Proposed accelerator based systems are similarly diverse with different proposals making different technology choices.

There are broadly two kinds of critical reactors that can be used for transmutation of nuclear waste: thermal neutron and fast neutron reactors. In all cases the fuel is configured such that it can sustain a nuclear chain reaction. The reaction is controlled by the use of neutron absorbers, including control rods that can be removed and inserted in order to change the reaction rate.[79]

Thermal-neutron critical reactors include conventional light-water reactors, which are the predominant type of reactor used worldwide.[80] The water acts as a moderator of the neutrons, resulting in a thermal neutron spectrum. They use ceramic oxide fuels which, in the once-through cycle in effect in the United States, consists of uranium oxide. The uranium is low-enriched uranium (~3-5% U-235). Specially designed reactors have also been fueled with mixed oxide fuel (MOX) consisting of a mixture of plutonium oxide and uranium oxide.[81] France has the most extensive civilian nuclear program using

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[77] Thermal neutrons can be characterized by their most probable velocity at room temperature (which corresponds to about 0.0253 eV). In reality, they have a distribution of velocities (and therefore energies). The characteristic temperature in a reactor is actually around 300^0C degrees rather than 20^0C (room temperature). However, that does not make a significant difference. See Bodansky 1996, p. 56.

[78] Bodansky 1996, pp. 61-62 and p. 64. It should be noted that this does not hold true for non-fissile actinides which will undergo neutron capture in a thermal spectrum, but can be fissioned in a fast spectrum. In fact, this is the definition of a "fissile material," it is an isotope which can undergo fission by a thermal neutron.

[79] Neutron absorbers are different than neutron moderators. A moderator only slows the neutron down whereas an absorber will actually absorb the neutron so that it no longer exists as a distinct particle but becomes part of the absorber's nucleus.

[80] Of the approximately 400 commercial nuclear power reactors worldwide, light water reactors (LWRs), in which ordinary water acts as both coolant and moderator, account for approximately two thirds. The other main types of reactors in commercial use include heavy water reactors (which use natural uranium oxide fuels and water with a high percentage of deuterium atoms which is an isotope of hydrogen), light water cooled graphite moderated reactors, and gas-cooled reactors.

[81] MOX fuel use is the most straightforward of transmutation schemes and the only one implemented so far. Reactor fuel is reprocessed to separate the uranium and plutonium, which are then fabricated into MOX

MOX, with 20 out of 58 reactors fueled with MOX.[82] In commercial operations, aqueous reprocessing has been used for light-water reactor fuel (generally based on PUREX). For more complete transmutation, further aqueous processing is required to separate the minor actinides, technetium, and iodine and those processes have not been developed to the industrial scale.[83] Pyro-processing has also been proposed for LWR fuel, but this technology is still at the laboratory stage (with some parts of the technology having been recently demonstrated on sodium-bonded spent nuclear fuel).[84]

The other class of critical reactors that could be used for transmutation is fast neutron reactors based upon technology developed for the breeder reactor program. As stated, they use a fast neutron spectrum (for the breeder program this was advantageous because it resulted in more neutrons per fission of plutonium which could go on to create plutonium in the breeder blanket). Fast breeder reactors can also use oxide fuels which are a combination of PuO_2-UO_2 (though with higher enrichment than thermal reactors). Other fuels have also been developed or proposed for breeder reactors, including the metal fuel for the Integral Fast Reactor, which forms the basis for some accelerator based transmutation proposals. Since fast reactors cannot use a moderator the coolant material must be such that it has a high atomic mass in order to minimize moderation. Liquid sodium has been the main choice. However, sodium has some drawbacks, not least of which is its chemical reactivity in air making leaks of the sodium coolant a particular hazard. Problems with the sodium coolant system have plagued breeder reactors such as Superphenix in France and the Monju reactor in Japan.[85]

Reactors can also be configured to operate in a sub-critical mode (i.e. the configuration of the fuel is such that a self-sustaining chain reaction cannot occur). However, this requires an additional source of neutrons for the nuclear reactions in order to sustain a chain reaction. Accelerator based transmutation would use accelerated protons to create neutrons through spallation reactions on a heavy target. These neutrons would enter the sub-critical reactors. Sub-critical reactors, which are still at the conceptual design stage, could use solid or liquid fuels and could have either a thermal or fast neutron spectrum. While all of the components of the ATW systems that appear most likely to be developed are based on existing technology, there would need to be significant development and scaling-up of the technology to meet the parameters necessary for ATW. An integration of different technologies that have never been part of a single process before also poses major design, testing, and safety challenges.

The choice between critical and sub-critical reactors and between thermal and fast neutron spectrum is a complicated one depending on many factors. For example, a

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fuel elements. However, the cycle can only be repeated a limited number of times due to changes in the composition of the plutonium and actinides which make the reprocessing more hazardous and affect reactor control and increase the cost of plutonium which has been irradiated multiple times. This is exacerbated by the fact that there are already large stocks of separated plutonium, making the multiple recycling of plutonium even less economical. See Chow and Jones 1999.

[82] CNE 2000, p. IX

[83] OECD-NEA 1999b, pp. 35-36.

[84] ATW Roadmap 1999d, p. 10 <<and NAS reviews of EMT>>

[85] Bodansky 1996, p. 87, pp. 96-97, and pp. 245-251. It should also be noted that thermal breeder reactor cycles have also been suggested but have made even less progress than fast breeder programs and were largely abandoned in the 1950s in favor of fast breeders.

thermal spectrum is more efficient at transmuting the long-lived fission products than a fast spectrum. On the other hand, non-fissile actinides fission more easily in a fast spectrum.

Waste Management

As will be discussed in more detail in Chapter V, there would be significant waste management issues remaining after all the current and projected stocks of spent fuel are transmuted. This is true even with the limiting assumption that nuclear power is phased out and that no new reactors are built or that transmutation reactors are not converted into a new nuclear energy industry with breeding of more fissile materials as has been proposed.

Residual actinides and long-lived fission products will have to be sent to a repository, as will those long-lived radionuclides that could not be transmuted. If not sent to the repository, the medium-lived fission products would have to be stored for upwards of 300 years or more. Additionally, there will be the waste from reprocessing operations.

A major open question is the fate of waste from prior reprocessing operations, both for military and commercial purposes. Some is still in storage tanks and some has been solidified, but none of this waste has been placed into a repository. This waste contains medium lived fission products, long-lived radionuclides and actinides (such as americium and curium). Some transmutation schemes would attempt to remove some radionuclides (particularly the minor actinides) from this waste, but it remains to be seen whether this is feasible, either technically or economically. In its review of transmutation, the National Academy of Sciences panel did not consider it a viable option for the military waste in the United States.[86] Thus all of this waste would still require disposal.

The uranium poses a particular problem. As discussed, the uranium in spent fuel will have to be separated with very little process losses in order to avoid production of new actinides by neutron capture. However, the question then becomes what to do with the uranium.

Proposals to handle long-lived fission products and uranium, either by burying them as low-level waste (after dilution) or re-using them in some manner, raise serious questions as to the health and environmental effects of transmutation. Either option would result in extremely long-lived radionuclides originally slated to be disposed of in a deep geologic repository instead being sent to shallow land burial. This is discussed further in Chapter V.

Overview of Proposed Transmutation Schemes

Transmutation is being actively researched in a number of countries. In particular, France has formalized its transmutation research as one branch of its

[86] NAS-NRC 1996, p. 9. The military high level waste contains much smaller amounts of transuranic radionuclides making it impractical to transmute these waste. The panel did recommend, however, some processing in order to facilitate disposal of the waste.

government-mandated examination of nuclear waste options.[87] Research is also underway in the United States, Russia, Switzerland, and Japan among others.[88] It is impossible to describe in detail in this report every research and development program. However, a number of the programs will be described in the following three chapters. Table 5 provides an overview of the characteristics of some of these transmutation proposals. Three types of reactors (light water reactors, fast reactors, and sub-critical reactors) and two types of reprocessing have been proposed. The table shows the type or types of reprocessing associated with each type of reactor and the radionuclides that would be candidates for transmutation. Most transmutation schemes would use a combination of reactors and associated reprocessing technologies. For example, in one scheme, light water reactors would be fueled with mixed oxide (MOX) fuel - that is, fuel made with plutonium extracted from low-enriched uranium spent fuel. The MOX spent fuel then would be reprocessed and the transuranic actinides would be extracted to fuel a fast neutron reactor (commonly called a breeder reactor). The fast reactor fuel would, in turn, be reprocessed and the remaining actinides would fuel a sub-critical accelerator driven reactor.

[87] LOI n° 91-1381 du 30 décembre 1991 relative aux recherches sur la gestion des déchets radioactifs.
[88] For example, a recent major conference on accelerator transmutation, International Conference on Accelerator Driven Transmutation Technologies and Applications, 3rd, Prague, Czech Republic, 1999, was held in the Czech Republic and several Czech researchers spelled out their country's program for transmutation. A previous conference, in 1996, was held in Sweden.

Table 5: Overview of Transmutation Schemes

Reactors and neutron sources	Type of Reprocessing and candidate radionuclides for transmutation	Comments
Light water reactors (LWRs) (the most common type of commercial nuclear reactor) The reactor is critical and fueled with either low-enriched uranium or mixed oxide uranium-plutonium fuel.	Reprocessing: aqueous Radionuclides: Primarily plutonium, Tc-99, I-129.	• Creates high proportion of higher mass actinides with associated severe radiation hazards • Reprocessing creates large amounts of liquid radioactive waste • Issues of reactor safety • Cannot fission most actinides • Heavy transuranic build-up, creating waste management problems
Fast reactors: The reactor is critical and can be fueled with plutonium, uranium or, potentially, fuel containing some minor actinides.	Reprocessing: mostly dry in advanced schemes. Radionuclides: Plutonium and possibly minor actinides. Tc-99 and I-129 may be possible but only in moderated targets outside the reactor core.	• The development of fast reactors has been crippled by persistent problems • Fission products are not efficiently transmuted • Heavy transuranic build-up though to a lesser extent than with LWRs • Issues of reactor safety
Sub-critical reactors: an accelerator-target system provides fast neutrons to a sub-critical reactor	Reprocessing: the reprocessing can be all aqueous or all dry or a combination of the two Radionuclides: plutonium and minor actinides. Tc-99 and I-129 may be possible but only in moderated targets outside the reactor core.	• Sub-critical reactors are only at the R&D stage • Cost is projected to be high. • Reactor safety still an issue • Fission products are not efficiently transmuted

Chapter II: Separation

No technology can selectively transmute the long-lived radionuclides of concern to a degree meaningful for waste management while they are contained in the spent fuel. They need to be extracted from the spent fuel and then separated from each other. There are three main reasons why such separation is necessary.

1. To be properly transmuted the radionuclides of concern have to be separated from the neutron absorbing elements[89], the build up of which in the fuel interferes with, and eventually stops, the nuclear reactions that are essential to transmutation.

2. The uranium has to be taken out prior to transmutation because it makes up most of the mass (typically about 94%) of reactor spent fuel. The transmutation of uranium for waste management would be prohibitively expensive. Moreover, the route of transmutation for the uranium-238 would be its transformation into plutonium-239. This would result in the build-up of one of the most important radionuclides that transmutation seeks to eliminate when it is used as a waste management technique. The build-up of plutonium stocks is rather a characteristic of those nuclear reactor schemes that seek to create a long-term energy future based on plutonium, as was the case for the worldwide breeder reactor program that, overall has been a technical failure and a financial loss.[90] The transmutation of uranium-238 would then require a two step process. First, the uranium-238 would be transmuted into plutonium-239, then the plutonium would be transmuted. This would create huge new amounts of long-lived fission products and transuranic elements and would require a transmutation system far larger, costlier, more polluting, and more dangerous than the ones that have been proposed. Basically it would mean relying on costly nuclear power for energy for the long-term.

[89] Most fission products created during the process of reactor operation are neutron absorbers. Some of these fission products belong to the lanthanide group of elements, which chemically resemble transuranics. This is an important complicating factor in transmutation systems.
[90] Makhijani 2000

3. Because some elements are a mixture of isotopes, the gain arising from the transmutation of a long-lived isotope into a short-lived or stable isotope can be offset by the simultaneous transformation of a stable or short-lived isotope into a long-lived isotope. Separation of isotopes is necessary to prevent this counterproductive result.

Separation is a costly process that creates additional wastes and radiation hazards. It also raises proliferation concerns, since the same techniques can be used to separate weapons usable materials from spent fuel.

Many hurdles still remain to obtain a very high degree of selective conversion of long-lived into short-lived radionuclides or stable elements and some of the principal ones relate to separations processes. The main radionuclides of concern in the spent fuel to long-term waste management are the:

- transuranics: the isotopes of plutonium, neptunium-237, and the isotopes of americium and curium,
- long-lived fission products: iodine-129, technetium-99, selenium-79, tin-126, palladium-107, and cesium-135,
- long-lived activation products: chlorine-36 and zirconium-93
- radioactive gases: medium-lived tritium and krypton-85, and carbon-14 dioxide (the long-lived activation product carbon-14, although not a gas in the fuel, is converted to CO_2 during reprocessing),
- "medium-lived" fission products: strontium-90 and cesium-137.

In an ideal transmutation program, almost all of the above radionuclides would be transmuted in short-lived radionuclides or stable elements. However, such a program is not practical for a variety of reasons, including:
(i) The neutron absorption cross section of some fission and activation products is very small, which makes transmutation very expensive,
(ii) There are several isotopes of some elements in spent fuel in the case of many radionuclides. In some cases, isotopes of widely varying half-lives of the same element are mixed in the spent fuel, making it impossible to transmute the long-lived isotopes into short-lived ones without triggering the reverse process.
(iii) It is very difficult to trap radionuclides present as gases or that are transformed into gases during reprocessing and put them into physical forms suitable for transmutation.

In view of these limitations, transmutation is, to some extent, feasible only for most transuranics and for only two of about a dozen long-lived and medium-lived fission and activation products. These two are the long-lived fission products iodine-129 and technetium-99.

Although the transmutation of the medium-lived strontium-90 and cesium-137 is not practical they are the main source of heat in the spent fuel. This thermal contribution requires more space for some deep repository scenarios. Therefore, their separation and storage above ground for several centuries has been proposed.

In general, a first separation will aim at separating the main actinides, namely uranium and plutonium and a few long-lived fission products from the rest of the fission products, which are neutron absorbers. For transmutation approaches that seek a very partial reduction of long-lived radionuclides, such separation into broad groups of elements is enough. For other approaches where more complete transmutation (within the limitations described above) is the goal, additional separation steps are needed. However, the lanthanides (also called rare earths) that contribute one-third of the total mass of fission products, are co-extracted with the actinides because these two groups of elements have similar chemical properties.

General description of separation processes

Although there are numerous separation processes, they belong to only two main categories: aqueous and dry. Aqueous processes are used for radionuclides in the form of oxides, dry processes have been developed and are used for radionuclides in the form of metals, but they can be modified for radionuclides in the form of oxides. In that case, the oxides are first reduced to metals.

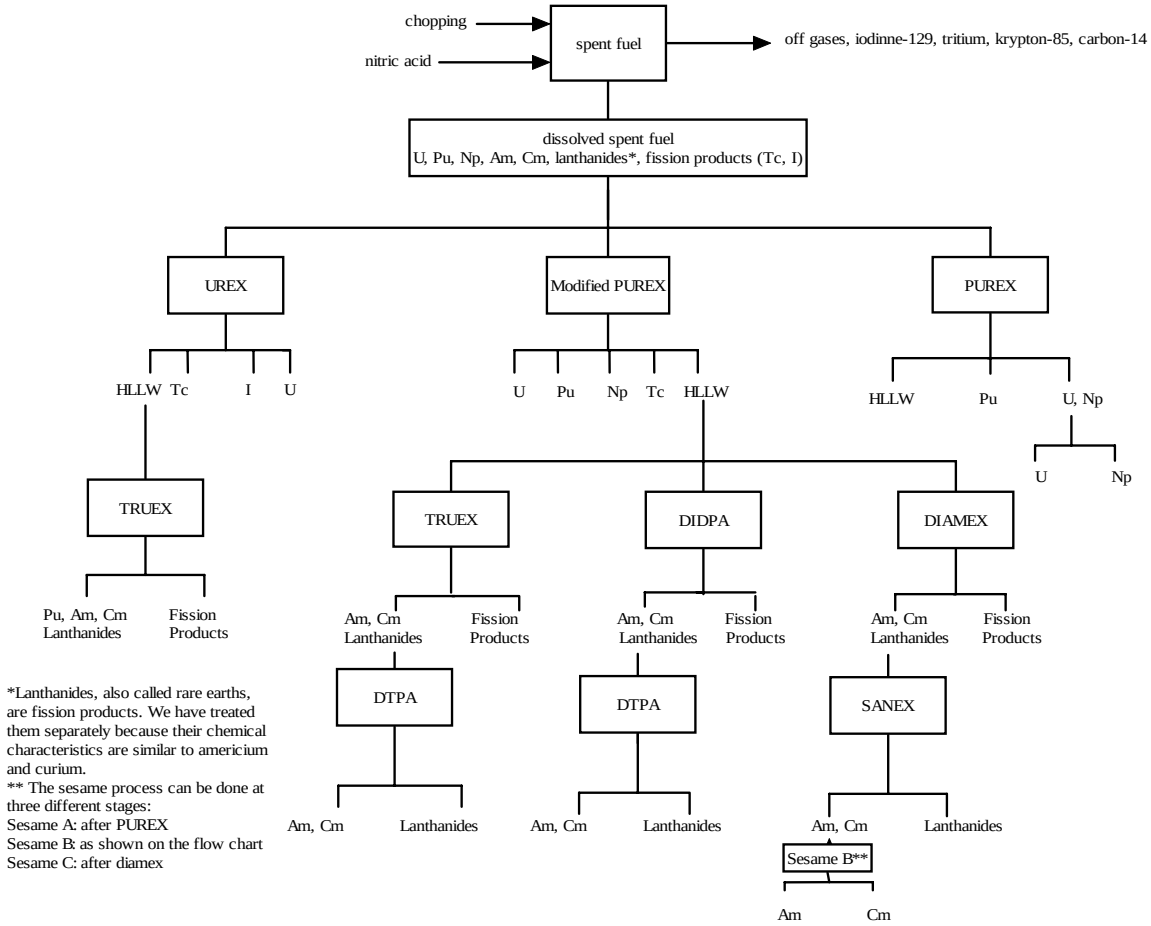
Aqueous processes rely on the preferential dissolution of elements under specific chemical conditions. In the process of dissolution in an acid the specific tendency of an element (and its chemical compounds) to react can be controlled by generating different oxidation states, which are described by numbers reflecting the amount of positive charge on an ion. The control of oxidation numbers allows the extraction of an element from a solution when an organic extractant, like tributylphosphate, is added.

Dry processes make use of electrolysis and the different chemical potential of each element. Given the right set of conditions, a metallic radionuclide is made to dissolve in a molten salt at its well-defined potential by applying the proper current. Once dissolved the radionuclide becomes a metallic ion. The current is then modified to allow for the transport of the radionuclide to a cathode where it is deposited as a metal. For the purpose of transmutation this principle can also be applied to groups of elements. The electrolytic process is variously called pyroprocessing or electrometallurgical processing.

Among the numerous processes in each of these categories, only a few have been used on an industrial scale for any length of time. Of these the PUREX (for Plutonium-Uranium EXtraction) process which uses nitric acid as a solvent is by far the best established and is currently the only one used on a large scale. Most other separation technologies are still in various stages of research and development. For reasons of space and clarity we will describe only those processes which are being researched in the countries which are seriously researching separation and transmutation as an option for the long-term management of nuclear wastes. These countries are the United States, Japan, and France.

The flow diagram in Figure 4 shows the various types of aqueous reprocessing that may be used to separate radionuclides prior to their insertion in a reactor for transmutation. The aqueous processes all have the dissolution of uranium oxide (UO_2) spent fuel from light water reactors (LWRs) as their starting point. Once the spent fuel is dissolved in nitric acid different levels of separation can be achieved, as currently with PUREX.

Figure 4: Overview of the Proposed Aqueous Processes



The main difference between the aqueous and dry processes resides in the fact that the aqueous processes are designed to possibly achieve a high degree of separation of transuranics from each other, whereas the dry processes are designed to extract the transuranics radionuclides as a group. This is the basis of the claim made by proponents of dry processes that they are proliferation-resistant since plutonium is not extracted by itself but in combination with other transuranics. (This is a misleading non-proliferation claim, as discussed in Chapter V.) Another important difference resides in the fact that the molten salts used in the dry process for dissolution are more radiation resistant than the organic extractants used in the aqueous processes. That is, the molten salts can withstand higher levels of radiation without being damaged by it. This allows for a shorter cooling time between the unloading of the spent fuel and the separation process.

Aqueous processes

The basic aqueous separation technology (also called reprocessing), for transmutation would be the same as that now used for commercial reprocessing, the objective of which is to separate the plutonium and the uranium from the fission products in the spent fuel and from each other. In principle, this allows recycling of uranium and plutonium in reactors. The plutonium is separated for use as a fuel, while most of the uranium, which

is non-fissile uranium-238, was supposed to be used as the raw material for conversion into plutonium in breeder reactors. PUREX is the only process that is currently being used on an industrial scale in such separation operations.

Separation for the purpose of waste transmutation, rather than as part of a long-term, reactor-based energy scheme, uses, as a first step, one of two aqueous processes: a modified PUREX process (similar to PUREX) or the UREX (URanium EXtraction) process. The latter process is also similar to the PUREX process, with the principal difference that plutonium is not separated but is retained in the high-level waste stream, which also contains almost all the fission products. Either of these processes provide only a crude level of separation, which would allow for very limited transmutation. Further separation following PUREX or UREX would be required for more complete transmutation. However, the processes that would be used to accomplish this, which constitute a veritable alphabet soup of acronyms, are still under development.

The PUREX process[91]

The PUREX process shown in Figure 5 has been used for several decades for the extraction of plutonium for military as well as commercial purposes. Today, it is used mainly for commercial purposes, though several countries are operating military reprocessing plants. Among them the US claims that this is necessary for environmental reasons because some of the spent fuel is corroding and releasing radioactive material in the cooling pools.[92] Russia is also operating two military reprocessing plants ostensibly for spent fuel management.

In the first step, the spent fuel rods are chopped up into short pieces. The contents of the fuel rods are dissolved in hot nitric acid and the empty hulls become part of reprocessing solid waste. However, some fission products, notably technetium, ruthenium, rhodium and palladium do not dissolve completely and settle to the bottom. They are removed by filtration and then incorporated in the vitrified or cemented wastes. During this first step, the dissolution of spent fuel results in the venting to the atmosphere of tritium, carbon-14 (as carbon-14 dioxide), krypton-85 and some iodine-129.[93] Most of the iodine-129 is released to the ocean and much of the rest to the atmosphere.[94] The widespread demand for the elimination of discharges of radioactive waste from reprocessing into the seas has given rise to the possibility that the amount of iodine-129 that would need to be transmuted may increase substantially.

After the dissolution step, the nitrate solution is exposed to the solvent tributylphosphate (TBP) which is mixed with kerosene to improve its physical properties. The TBP selectively separates the plutonium and the uranium from the rest of the solution. During

[91] For a detailed description of the PUREX process see Benedict, Pigford and Levi 1981, pp. 466-514
[92] Sachs 1996, p. 1
[93] COGEMA 1997, pp. 11-12
[94] CNE 2000, pp.45-46. At the La Hague French reprocessing plant, 97 % of the iodine is released to the sea after being trapped, 1 to 2 % are being trapped by trapped by solid filters and the rest is being released to the atmosphere (1%) and fixed in the cladding (0.2 to 0.3 %).

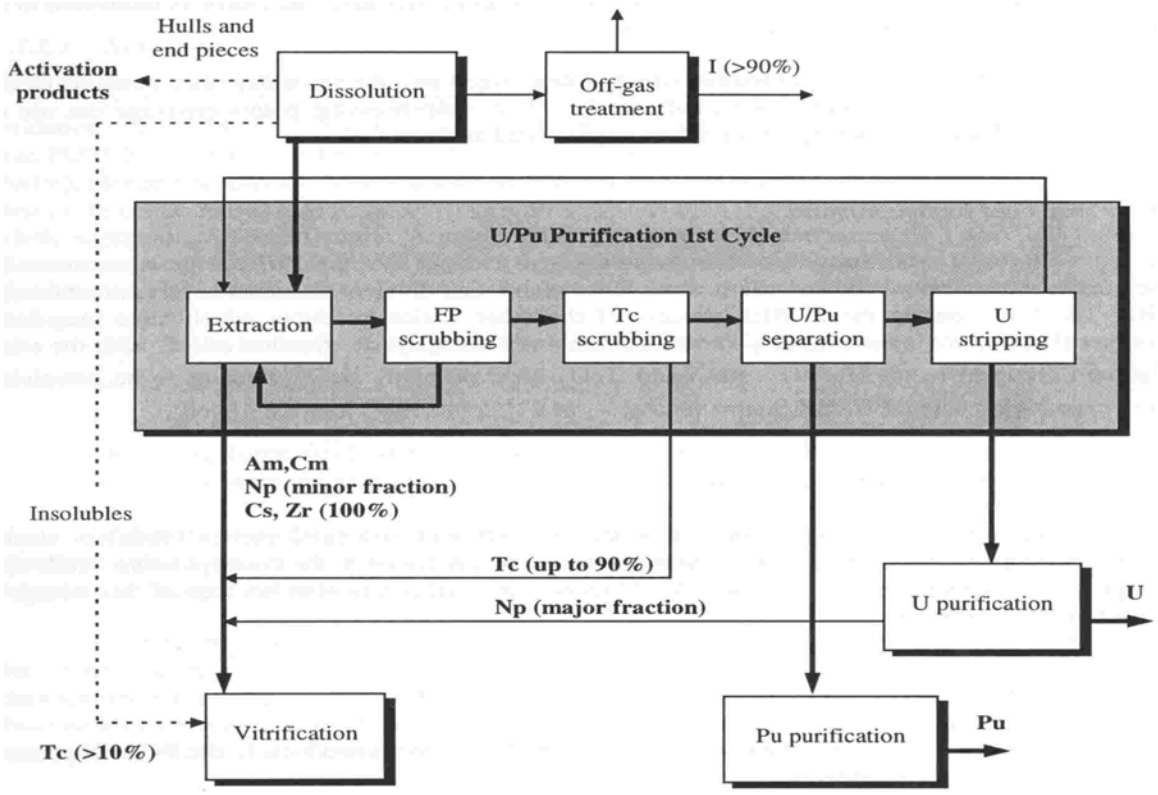
that process some fission products, such as technetium-99, are also extracted by the TBP.[95] The majority, 99% to 99.9%, of the fission products and americium and curium remains in the nitrate solution. Some of the neptunium goes with the plutonium and uranium and the rest remains with the fission products.

The next step is the separation of plutonium and uranium from each other. The neptunium remains mainly with the uranium.

The remaining step is the purification of plutonium and uranium; in particular the removal of neptunium-237 from uranium. In the commercial reprocessing plants, the high level liquid waste containing the fission products is typically calcined (dried and converted to oxide) and then vitrified along with undissolved residues, the traces of plutonium and the neptunium separated from the uranium.[96]

The amount of liquid high-level waste resulting from this process is 5,000 liters per metric ton of heavy metal.[97] Far larger amounts of liquid low-level radioactive wastes are created and discharged into the environment.

Figure 5: PUREX Process (An example of UP3 La Hague)



Source: Reprinted with permission from OECD-NEA 1999b, p. 115

[95] OECD NEA 1999 b, p 114.The rest of the technetium-99 (10 to 20 %) stays in the nitric acid solution along with ruthenium, rhodium and palladium as insoluble residues and is incorporated in the vitrified or cemented wastes.
[96] CNE 1998, p. 49
[97] OECD-NEA 1999b, p.122

The UREX process[98]

This process would extract uranium from LWR spent fuel along with technetium-99 and neptunium-237. The uranium is then separated from the other two radionuclides. The trapping of iodine-129 from the off-gas, its dissolution in the aqueous solution and its recovery from it is also proposed. The UREX process is an alteration of the PUREX process. The main difference is that the plutonium is rejected, along with the americium and the curium, to the high-level liquid waste.

The main objective of the UREX process is to extract uranium containing very little neptunium-237 and technetium-99. According to Argonne National Laboratory, this would allow the separated uranium to meet the US criteria for Class C "low-level" waste, which is allowed to be disposed of in shallow land burial sites.[99] Such waste cannot be so disposed in most other countries. In Europe, it is called "intermediate-level" waste and deep disposal is required. Because uranium represents about 94 % of the weight of the heavy metal in spent fuel, this approach could drastically reduce the space necessary for a deep geological repository. However, it is environmentally unsound in our view (see Chapter V).[100]

Because PUREX is a well-established industrial process, the setting up of the UREX process can be done largely with existing technology. However, the main modification required is quite significant, since it involves the development of a method to safely leave the plutonium in the high-level liquid waste. The management of such waste will pose new challenges, such as criticality issues, to a far greater extent than with the management of high-level waste from the PUREX process. Some other technological barriers will have to be addressed:

- There could be an increase in the volume of waste due to the addition of chemicals to keep the plutonium with the fission product stream.
- The neptunium-237 and technetium-99 which are initially co-extracted with the uranium need to be separated from the uranium with a high degree of decontamination, in order to produce a nearly pure stream of uranium.
- The iodine-129 will need to be removed from the off-gas stream in a form that can be easily manufactured into targets and with little loss to the environment.
- The amount of undissolved technetium-99 needs to be minimized.

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[98] Based on ATW Roadmap 1999d, pp. 5-14

[99] ATW roadmap 1999d, p.5

[100] The status of various forms of uranium that may have to be disposed of as waste is very murky, at least in the United States. In the context of a licensing proceeding for a uranium enrichment plant, the NRC declared that depleted uranium could be disposed of a Class A radioactive waste. The Argonne document cited above would treat uranium recovered as part of the UREX process as Class C waste, even though both forms of uranium are alpha-emitters with specific activities well above 100 nanocuries per gram. In our analysis this makes separated uranium s well as depleted uranium analogous to transuranic waste, therefore requiring repository disposal.

The modified PUREX process[101]

The modified PUREX process allows for the extraction of almost all of the neptunium along with the plutonium and the uranium. After dissolution of the spent fuel in nitric acid neptunium is present in two ionic forms, NpO_2^+ and NpO_2^{2+}. TBP does not have any marked affinity for the first ion, but does for the second one. It is therefore possible to extract neptunium by adjusting the solution so that NpO_2^{2+} becomes essentially the only species. Research underway indicates that such a process is feasible.[102]

After its coextraction with plutonium and uranium, neptunium-237 can be selectively separated using the same PUREX cycle or specific reagents such as butyraldehyde. The PUREX process can also be adapted to separate the technetium-99 that is extracted - along with the uranium and the plutonium - from the high-level liquid waste. Experiments indicate that 97 to 98% of technetium-99 and 99 % of neptunium-237 can be recovered from the high level liquid waste.

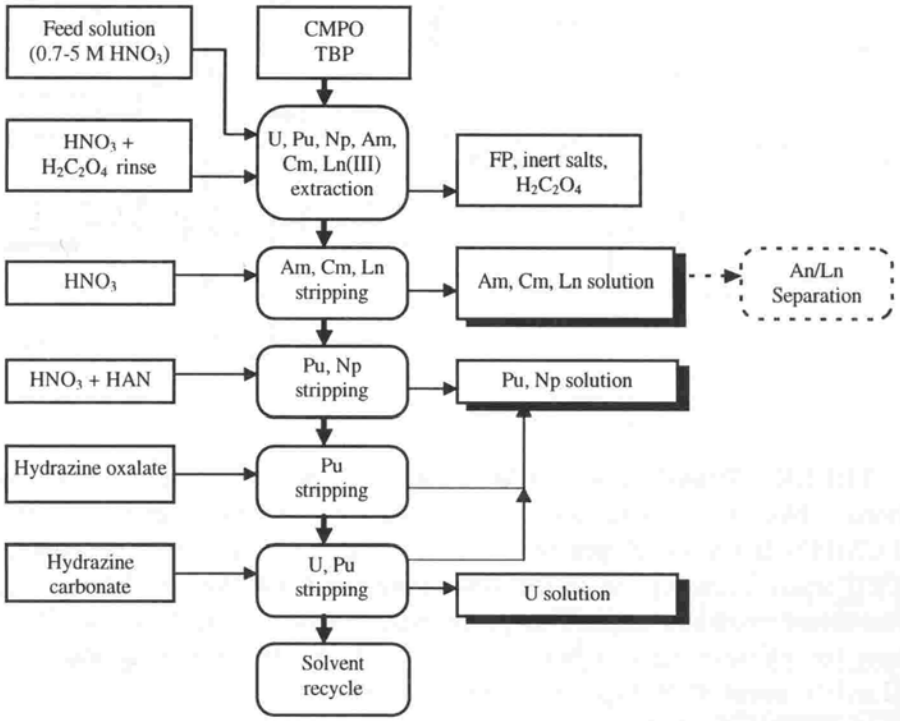
At this level of separation, plutonium, neptunium, and technetium can be transmuted, leaving a number of other problem radionuclides to be dealt with. Additional levels of separation are required for the transmutation of americium, curium. These experimental processes are called by their acronyms, TRUEX, DIDPA and DIAMEX.[103]. They are alternative approaches to further separation of radionuclides following the modified PUREX process (see Figure 4). Figures 3, 4, and 5 situate these three processes in their overall-reprocessing scheme. We provide a brief description of each of these three processes below. Because the lanthanides have chemical properties similar to the chemical properties of americium and curium the TRUEX, DIDPA and DIAMEX processes also extract them. The presence of the lanthanides is a nuisance because they are neutron absorbers and would interfere with the transmutation process.

The TRUEX process

The TRUEX (TRansUranic EXtraction) process shown in Figure 3 was developed at the Argonne National Laboratory in the 1980s to decontaminate the vast quantities of transuranic wastes originating from Cold War production of plutonium materials for nuclear weapons.[104] This process aims at separating americium and curium from high-level liquid waste. It requires PUREX or the modified PUREX process as its front end. The TRUEX sequence makes use of an organophosphorous extractant, CMPO (carbamoylmethyl-phosphine-oxide) which has a high affinity for americium, curium and the lanthanides. The TRUEX sequence would contribute an additional 750 liters of high-level liquid waste per metric ton of heavy metal to the 5,000 liters of high-level waste from PUREX.[105]

[101] Based on OECD-NEA 1999b, p.117
[102] Boullis 1997, p. 86
[103] Based on OECD-NEA 1999b, pp. 119,121, 124
[104] OECD-NEA 1999b, p.121
[105] OECD-NEA 1999b, p. 122

Figure 6: TRUEX process



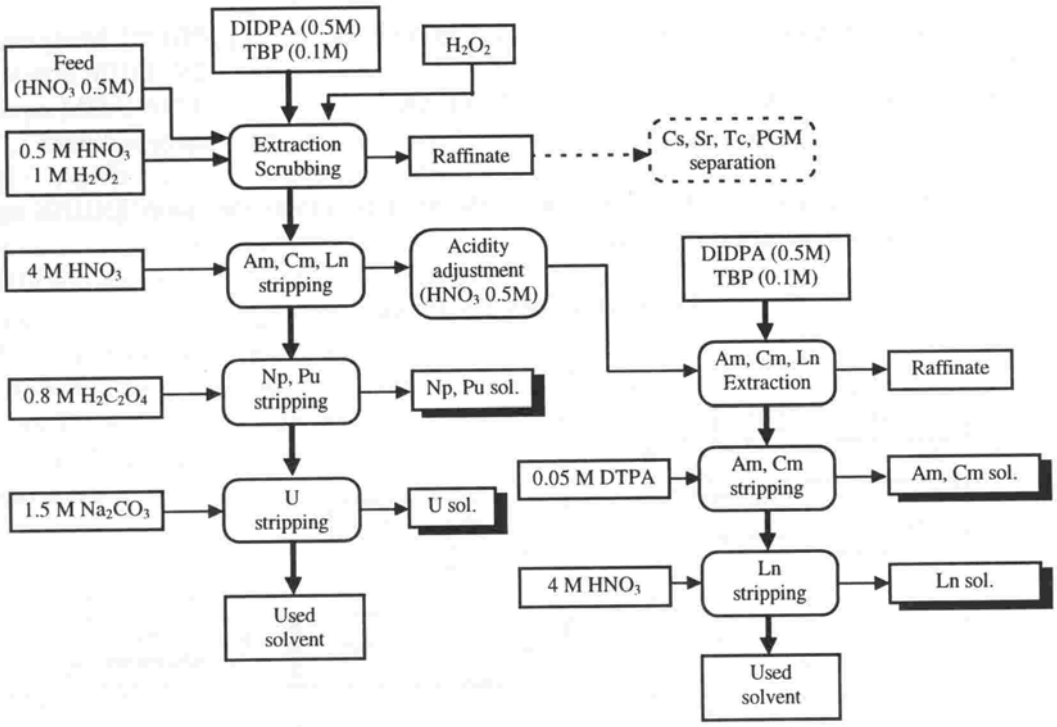
Source: Reprinted with permission from OECD-NEA 1999b, p. 122

The DIDPA process

After the high level liquid waste is taken through denitrification and filtration, the DIDPA process, shown in Figure 7, allows for the co-extraction of americium, curium and the lanthanides by an organic extractant (di-isodecylphosphoric acid -- hence the name DIDPA). Recovery levels of 99.99 % for americium and curium with real high level liquid waste have been shown to be technically feasible on an experimental basis, according to Japanese researchers.[106] From the high level liquid waste cesium and strontium-90 would also be extracted with inorganic ion exchangers.[107] The amount of additional high-level liquid waste is on the same order as for the TRUEX process.[108] Further separation of americium and curium following the DIDPA process can be accomplished by selectively stripping them out of the organic phase into an aqueous phase by the use of an alcohol-carboxylic acid and DTPA (diethylenetriaminopentaacetic acid) to separate them from the lanthanides.[109] Figure 7 situates DTPA in the overall DIDPA process scheme

[106] OECD-NEA 1999b, p. 120
[107] OECD-NEA 1999b, p. 256
[108] OECD-NEA 1999b, p. 120
[109] OECD-NEA 1999b, p. 120

Figure 7: DIDPA process



Source: Reprinted with permission from OECD-NEA 1999b, p. 121

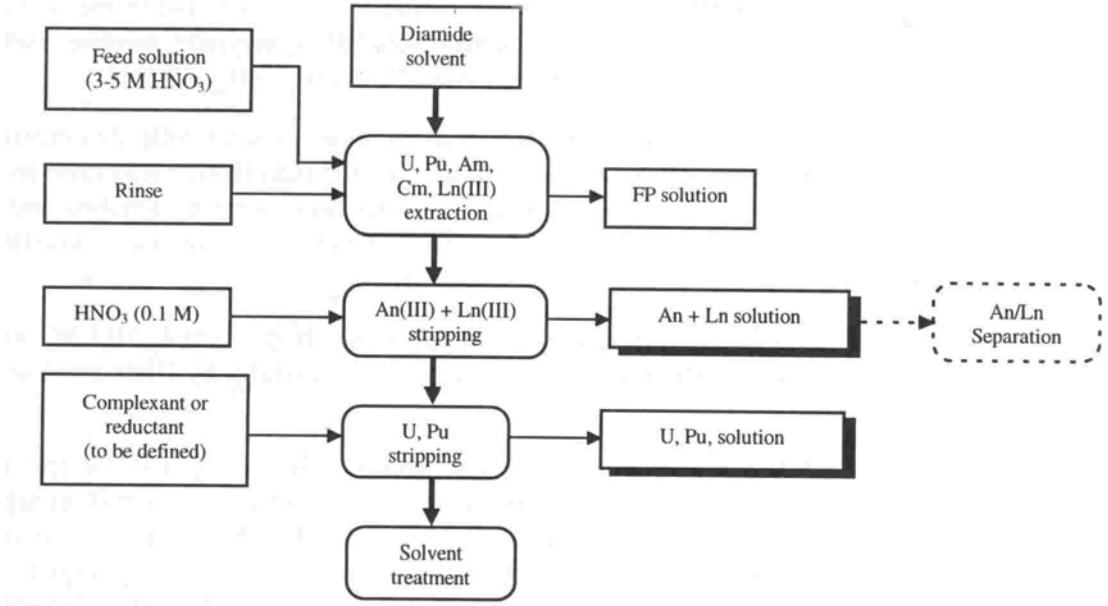
The DIAMEX process

The DIAMEX (DIAMide EXtraction) process shown in Figure 8 follows the PUREX process. DIAMEX extracts the lanthanides, the americium and the curium from the high level liquid waste.

The feasibility of this process has been demonstrated only at the laboratory level.[110] Since the extracting chemical di-methyl-di-butyltetradecylmalonamide (DMDBTDMA) is made up of oxygen, nitrogen, carbon and hydrogen, it has been proposed to incinerate it in order to minimize the amount of additional waste produced. However, incineration of radioactively contaminated materials poses its own problems and is so controversial that it has even been opposed by a task force of the Lawrence Livermore National Laboratory.[111]

[110] Boullis 1997, p.88
[111] Mendelsohn et al. 1990, p. 1

Figure 8: DIAMEX process



Source: Reprinted with permission from OECD-NEA 1999b, p. 125

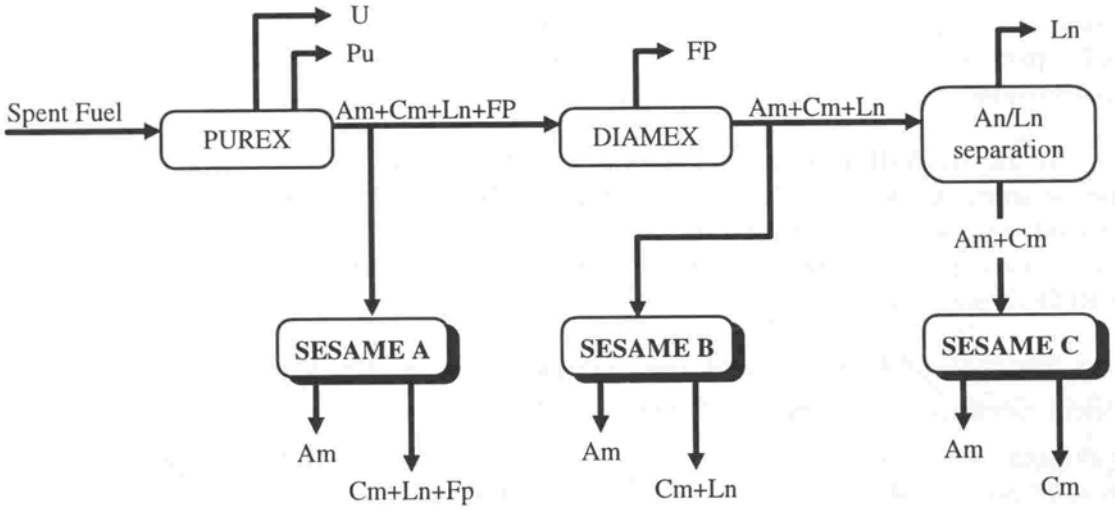
The SANEX process accomplishes the further separation of americium and curium from lanthanides following the DIAMEX process. This process uses a molecule made up of nitrogen, carbon and hydrogen (called nPr-BTP). [112] The atoms of nitrogen on that molecule bind selectively with the americium and curium allowing their separation from the lanthanides. Basic research on this process is being conducted at the laboratory level. The separation of americium from other radionuclide, that go by the acronym SESAME (Séparation Extraction Sélective de l'Américium par des Moyens Electrochimiques), is proposed to be accomplished at several alternative stages of separation .[113] The three possible separation schemes are shown in Figure 6. This process is based on changing the oxidation number of americium from III to IV or VI in nitric acid, followed by its selective extraction. The French are developing this process and the CEA (Commissariat à l'Energie Atomique) hopes that a separation technique will be available by 2006.

The various alternatives for the SESAME process that have been proposed would deal with the radionuclides associated with americium, notably curium and the lanthanides in different ways. The basic idea of separating americium from curium is to make the transmutation of americium more efficient.

Figure 9 and Figure 10 give an overview of the ways in which France and Japan have considered the use of aqueous process for their waste transmutation programs.

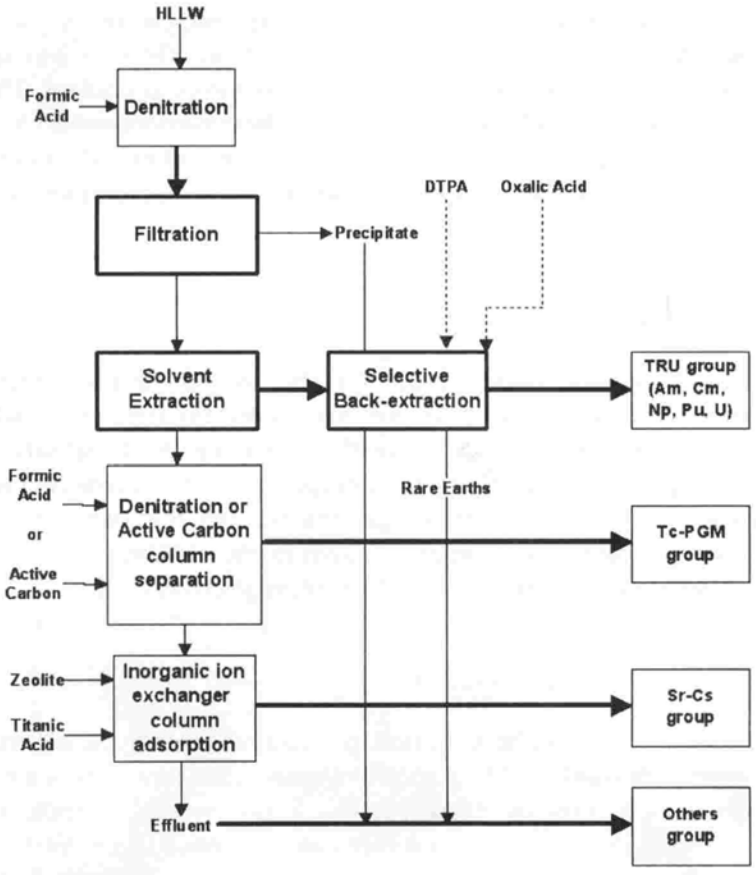
[112] CNE 1999, p. 61
[113] OECD NEA 1999b, p. 127

Figure 9: Aqueous scheme for France with Possible Separation Schemes Related to the SESAME Process



Source: Reprinted with permission from OECD-NEA 1999b, 128

Figure 10: Aqueous scheme for Japan



Source: Reprinted with permission from OECD-NEA 1999b, p. 257

Dry Processes

The objective of the dry processes is to separate the transuranics from the LWR, fast reactor, or accelerator spent fuel. There are broadly two types of dry processes. The difference between them resides not in the type of separation achieved (which is similar) but in the kinds and quantities of fuel that would be reprocessed.

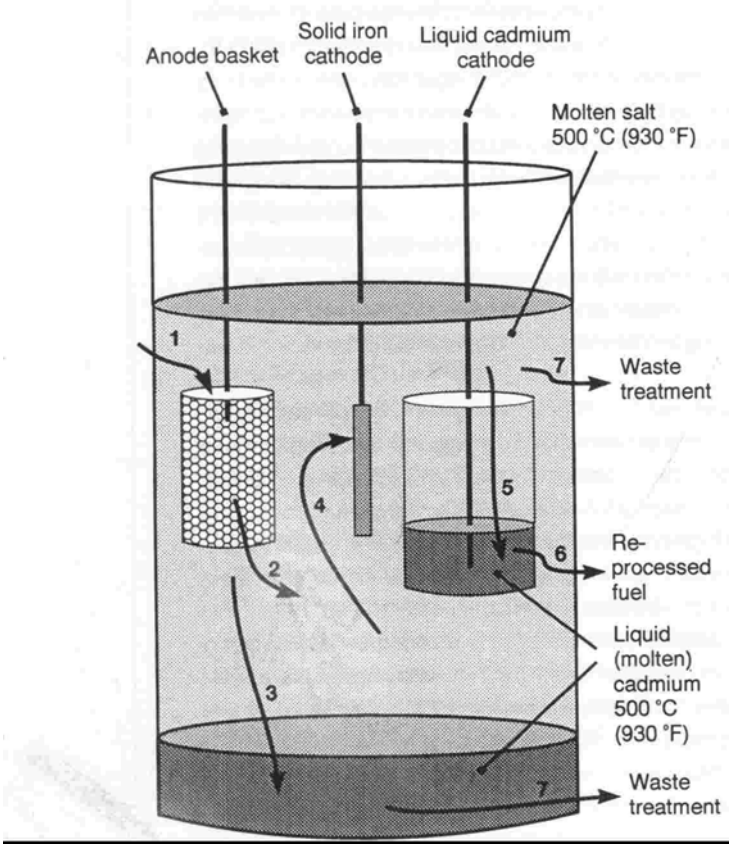
The core of the dry process is electrolytic separation of elements. This process, is variously named as "pyroprocessing", "pyrometallurgical processing" "pyrochemical processing" "electrorefining processing and electrometallurgical processing". It was developed by Argonne National Laboratory for the Integral Fast Breeder (ANL-IFR). The Integral Fast Breeder was canceled in 1994 but development of the dry process has continued, ostensibly for waste management purposes.[114]

The basis of pyroprocessing is that, given the right set of conditions, elements are converted into charged particles called ions when well-defined voltage is applied. The reverse process can also be made to occur electrically. This allows a selective separation of groups of elements by electrolysis.

Figure 11 shows a diagram of the process. Chopped spent fuel elements are dissolved at the anode in a solvent of molten salt (lithium chloride or potassium chloride) at 500^{o}C and a current is passed through the melt. The bulk of the uranium is deposited on a solid steel cathode while the transuranics, the remaining uranium and some lanthanide fission products are deposited on a liquid cadmium cathode. The major fission products, such as strontium and cesium remain in the molten salt from which they are removed. The two cathodes are then taken out of the solution and heated at 1000^{o}C to 1200^{o}C to remove the salts and the cadmium. The metals on the two cathodes are manufactured into uranium and transuranics/lanthanides ingots.[115]

[114] Sachs 1996, pp. 33-35
[115] Sachs 1996, p. 35

Figure 11: Diagram of the ANL-IFR Separation



Source: OTA 1994, p. 21

The dry process has been proposed for reprocessing ATW (Accelerator Transmutation of Waste) fuel and fast reactor spent fuel. In the case of ATW spent fuel, a chloride volatility process has also been proposed as an additional front-end step because it is well suited for the removal of the zirconium from the spent fuel.[116] Zirconium constitutes most of the weight of the fuel (being a filler used to limit the amount of fissile material in the reactor core), but does not fission.[117] Once the zirconium is removed, the rest of the material can be electrolyzed, as discussed above. The iodine-129 can also be removed from the molten salt and fabricated into targets. The technetium-99 would be incorporated into the fresh fuel rods for transmutation.

The basic electrolytic process works if the element to be separated is in metallic form. If it is some other form, an oxide, for instance, as is the case with LWR spent fuel, it must first be transformed into a metallic state.[118]

The front-end process for oxide fuel (that is, the process prior to electrolysis) is its reduction to a metallic form. Lithium chloride can be used to reduce oxide-spent fuel into a metal. It has the potential advantage of keeping down the amount of waste,

[116] ATW Roadmap 1999d. p. 17
[117] The proposed fuel cited in the ATW roadmap for ATW is 23 wt. % TRU and 77 wt. % zirconium.
[118] OECD-NEA 1999b, p. 135

because the resulting formation of lithium oxide can, in principle, be electrolytically decomposed and the lithium recycled. Once this is done the remaining steps are basically the same as the ones described above. A similar process is even proposed for the separation of transuranic elements from high level liquid waste. In this case an additional step of precipitating the radionuclides as oxides to take them out of the liquid is necessary.[119] Once the radionuclides are oxidized, the steps are the same as with oxide fuel.

Nitride fuel containing high concentrations of minor actinides has been proposed for reactors dedicated to their transmutation because such fuels may contribute to reactor safety margin and are, in principle, compatible with pyroprocessing. The technique is essentially the same as for metal fuel. During that process the highly enriched nitrogen-15[120] is recovered and recycled.[121]

Table 6: Various reprocessing processes associated with proposed transmutation schemes

Process	Country	Purpose	Status	Waste
Aqueous (oxide fuels only)				
PUREX	France, Japan	Plutonium and uranium extraction	Commercial in France[1]	5000 liters of high-level waste per ton of heavy metal
UREX	USA	Uranium extraction along with technetium-99 and iodine-129	Would need little R&D to get to the industrial level[2]	Similar to PUREX with minimum added waste. Most added reagent would be recycled
Modified PUREX	France, Japan	Uranium, plutonium, neptunium-237, and technetium-99 extraction	Laboratory level	No additional waste
TRUEX	USA, Japan	Americium, curium, and lanthanides extraction	Developed in the US. Laboratory level. Needs more R&D before pilot scale	750 additional liters of high level waste per ton of heavy metal. Solvent non-recyclable
DIDPA	Japan	Americium, curium, and lanthanides extraction	Laboratory level	Same order as TRUEX, but the solvent is recyclable
DIAMEX	France, investigated in Japan and US	Americium, curium, and lanthanides extraction	Laboratory level	No additional secondary solid waste expected
SANEX	France	Americium and curium extraction	Laboratory level	no data available
SESAME	France	Americium extraction	Laboratory level	no data available

[119] OECD-NEA 1999b, p. 136

[120] Nitrogen-15, rather than nitrogen-14, the most abundant isotope, is being used to avoid the production of carbon-14, an activation product arising from nitrogen-14.

[121] OECD-NEA 1999b, p. 137

Dry (metal and oxide fuels)				
Metal fuel	Country	Purpose	Status	Waste
1. ANL (IFR spent fuel)	USA, Japan	extraction of transuranics	Pilot scale	Small amounts of high-level salt waste
2. ATW spent fuel	USA	extraction of transuranics	Laboratory scale[3]	Small amounts of high-level salt waste
Oxide fuel				
1. ANL	USA, Japan	extraction of transuranics	Laboratory scale	Small amounts of high-level salt waste
2. Following PUREX	Japan	extraction of transuranics	Small scale experiment on simulated waste	Minimal, the chemicals used in the process are recycled
3. Following UREX	USA	extraction of transuranics	Laboratory and engineering scale experiments have been done[4]	Small amounts of salt waste

Notes

[1] France reprocesses 850 metric tons per year (out of its 1200 metric tons annually discharged) of its own domestic spent fuel. Of the remaining 350 metric tons 135 are contributed by MOX.[122]
Japan has signed contracts with Cogema and BNFL for the reprocessing of 7,000 metric tons of spent fuel. The Tokai-Mura reprocessing plant in Japan has a capacity of 90 metric tons per year. It is slated to be shut down in 2003 at which time the Rokkasho-Mura plant, with a capacity of 800 metric tons per year is schedule to start.

[2] The target is a facility with an annual capacity of 1,440 metric tons of heavy metal.[123] A pilot scale demonstration facility for the separation of 100 kg of TRUs is envisaged by the year 2015 to be scaled up to 1200 kg by 2018. This separation of 1200 kg of TRUs would correspond to the reprocessing of 130 metric tons of commercial spent fuel, about one tenth of the proposed amount 1,440 metric tons to be reprocessed.[124]

[3] The eventual goal of the industry is to reprocess 100 to 200 kg of ATW spent fuel per day with a recovery efficiency greater than 99.9% for transuranics and greater than 95% for technetium-99 and iodine-129. This is to be achieved by first a lab scale program handling 1 to 10 kg of ATW fuel per day followed by a pilot scale program with 10 to 25 kg of ATW fuel per day. Eventually a demonstration facility that would process 13 metric tons per year is envisaged. [125]

[4] The goal is to produce a design of a demonstration plant.[126] Engineering scale experiments on 5 to 20 kg**,** have been done to investigate issues associated with the process and the scaling up to 10 metric tons of fuel per year. Conceptual design for this last project has been done. It remains to study the separation of technetium-99 during that process.[127]

[122] Schapira 1997, pp. 19, 20.
[123] ATW Roadmap 1999d, p.10
[124] ATW Roadmap 1999d, p. 14-15
[125] ATW Roadmap 1999d, p. 23
[126] ATW Roadmap 1999d, p.12
[127] ATW Roadmap 1999d, p. 10

Overview of the programs by countries

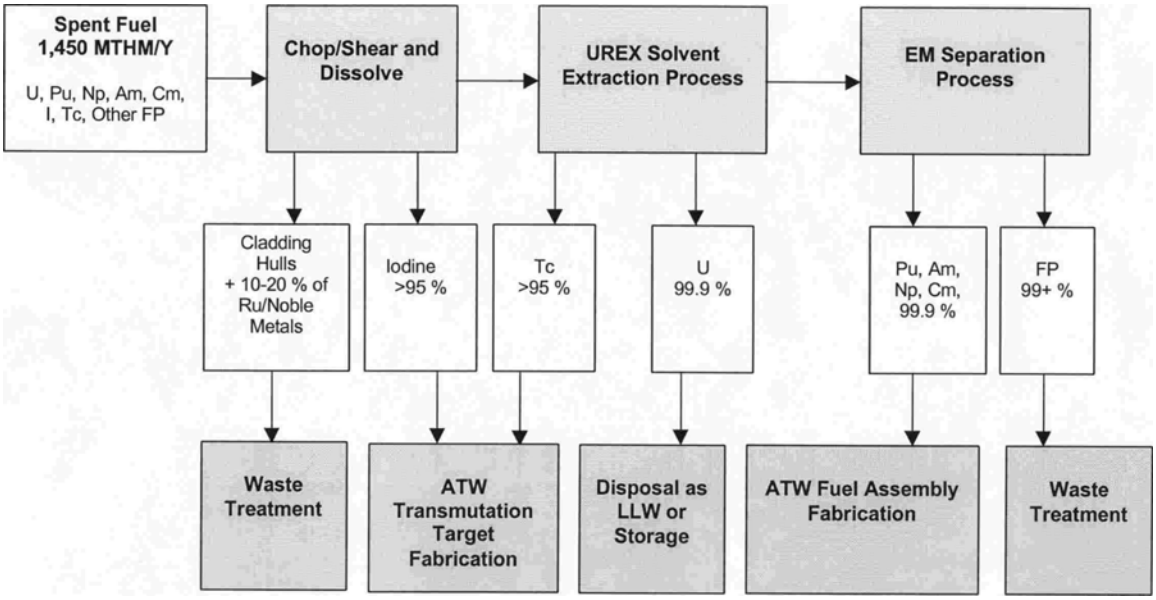
In Japan, and perhaps France (in one of the possible scenarios), the basic idea is to implement a two level fuel cycle which combines a reprocessing fuel cycle (devoted to electricity production) with an advanced fuel cycle (devoted to waste management). In the reprocessing fuel cycle, the spent fuel is reprocessed and the extracted plutonium is reused in the form of MOX in LWRs and fast reactors. For the advanced fuel cycle, the americium and the curium and some fission products are extracted from the high-level liquid waste and transmuted in fast or subcritical reactors.

Currently, the United States is committed to a once through cycle in light water reactors. But in a possible move away from this commitment, the DOE is researching transmutation in ATWs. The LWR fuel would be reprocessed and the radionuclides of concern transmuted during several passes through an accelerator with reprocessing occurring after each pass. The description of the program is given in a September 1999 report entitled "A Roadmap for Developing ATW Technology: Separations and Waste Forms Technology" by Argonne National Laboratory. In this report four separation schemes are described: a baseline process (the preferred option and three other processes.

1. In the baseline process (shown in Figure 12), commercial LWR spent fuel would be put through the UREX process. This would be followed by a dry process, which would separate the transuranics, in a metal form, from the fission products. The uranium stream would be disposed of in shallow landfills as Class C waste, the iodine-129 and technetium-99 would be fabricated into targets and the transuranics would be fabricated into fuel. The targets and the fuel would then be placed in a subcritical reactor for several cycles of transmutation and pyrometallurgical separation until the radionuclides of concern are destroyed. The entire cycle, from reprocessing LWRs spent fuel to multi-recycling in ATWs is shown in Figure 13. The program as currently envisioned by several DOE laboratories, including Argonne National Laboratory and Los Alamos National Laboratory, would process 1440 tons of commercial LWR spent fuel per year.
2. The second option would be an all-pyroprocessing process
3. The third options would be an all-aqueous process
4. The last process would consist of the UREX process followed by the TRUEX process to separate the transuranics from the fission products followed by the pyrometallurgical process to convert the transuranics from oxides to metals.

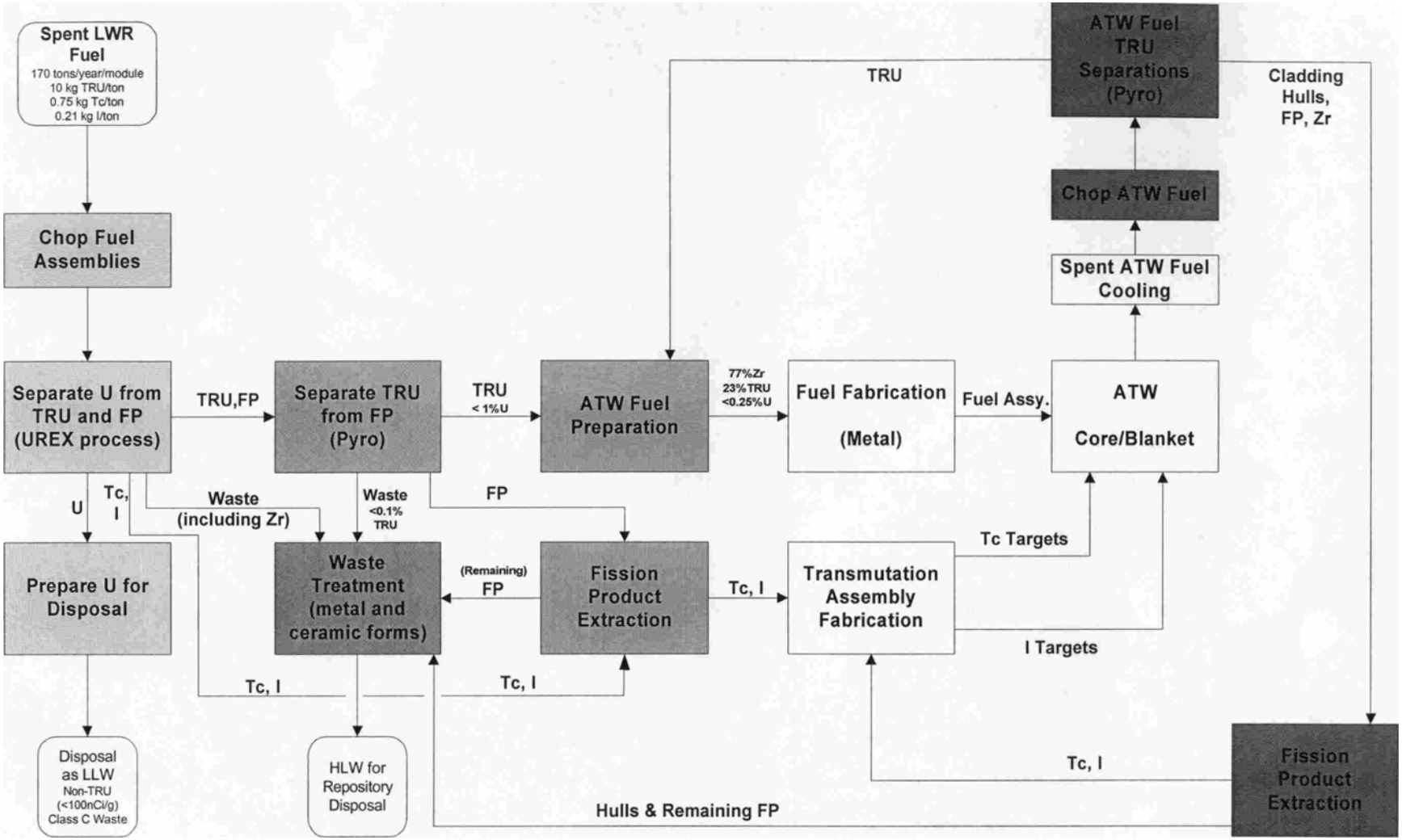
Although none of the processes mentioned above result in the separation of pure plutonium and the fabrication of civilian MOX fuel is not proposed, the construction of a UREX plant could be easily modified to allow for the extraction of pure plutonium.

Figure 12: Baseline LWR Reprocessing for the United States



Source: ATW Roadmap 1999d, p. 6

Figure 13: LWR and ATW Reprocessing Combined



Source: ATW Roadmap 1999d, p. 3

Japan[128]

Japan, as a matter of national energy policy, is committed to program of plutonium use in fast reactors. But it is also examining a program of accelerator driven reactors in a program called OMEGA (Options Making Extra Gains from Actinides and fission products).

Japan's nuclear fuel policy is to reprocess all of 1000 metric tons of spent fuel that are discharged annually from its LWRs. Currently the plan is to use the extracted plutonium as MOX in LWRs.

Japan sends most of its spent fuel to France and England for reprocessing. (It also does a little reprocessing domestically and plans to build a commercial plant by about 2005. France and England are also the countries where the plutonium is fabricated into MOX fuel. However, the loading of MOX fuel in Japanese reactors has been put on hold in the wake of public discontent after the September 30th, 1999 criticality accident in its Tokai-Mura plant and the report of falsification of MOX pellet data by BNFL (British Nuclear Fuels Limited), the manufacturer. As a result of the scandals and the prior breeder reactor accident in Monju in 1995, the future of Japan's plan for plutonium fuel use either in breeder reactors or as MOX in LWRs is very uncertain.

Japan is studying various separation processes that could be adapted to its transmutation program. These processes include two aqueous separations systems subsequent to PUREX - one based on the DIDPA process, the other on the TRUEX process. Japan is investigating dry processes as adapted to oxide fuels in the manner described in the section on this subject above.

For the advanced cycle – that is, breeders and ATW combined – Japan s studying the pyrochemical reprocessing of the various nitride, metal and oxide fuels that may be used in the various proposed reactors

France[129]

France, like Japan, is committed to a separation and transmutation program (refer to Chapter I) under the general rubric of Separation and Incineration. PUREX is used to extract plutonium, which is then fabricated into MOX. France is currently the world's largest user by far of MOX fuel in LWRs. This MOX fuel use is actually being carried out in LWRs in a once through manner, that is, after irradiation in the nuclear power plant, the MOX fuel is not further reprocessed.

Currently, with PUREX, France is annually reprocessing 850 out of 1200 metric tons of its own domestically produced spent fuel and is fabricating 115 tons of MOX fuel annually in its Melox plant at Marcoule. Most of the MOX fuel fabricated at Marcoule is

[128] For more information see OECD-NEA 1999b, Annex B
[129] For more information see OECD-NEA 1999b, Annex C

for consumption in French LWRs, a small proportion is for Japanese LWRs, though, as noted, Japan's program is stalled.

France also proposes a more advanced separation of the minor actinides and some fission products, using aqueous processes. These radionuclides would then be transmuted in fast reactors or sub-critical reactors. The separation processes would be the PUREX process followed by DIAMEX, SANEX, and SESAME. If the combination of all this processes is successful then the end product will be the separation of almost all the transuranics from each other. The research into separation is conducted by the CEA.

Health, safety and cost issues associated with separation

Currently the two main reprocessing activities are conducted at the Sellafield and La Hague reprocessing plants situated in England and France respectively. These two plants are discharging millions of curies in the air, the Irish Sea (Sellafield), and the English Channel (La Hague). The pollution in the sea affects the water from which people draw some of their food not only close to the site but also far away. This has prompted Ireland, Norway, Iceland and Denmark to demand that Sellafield and La Hague eliminate their so-called "low level" radioactive discharges. The implementation of a transmutation program relying on aqueous methods would greatly increase the amount of radioactivity discharged to the environment.

The PUREX plant at La Hague could be modified to accommodate the separation of neptunium. Cogema estimates that these modifications could be done in the next 10 years.[130] However the separation of americium and curium would necessitate the construction of new facilities with proper shielding. It is estimated that the cost of a pilot lab would be FF 450 millions (about $ 64 million). The initial investment for the separation of minor actinides and fission products could reach 5 billion francs (about 700 million US dollars) and this would not include the cost of running the plant.[131] These cost estimations are only for uranium oxide fuel that has been irradiated once. As discussed in Chapters III and IV, it is proposed to pass the radionuclides several times through a reactor, since only a modest amount of transmutation can be accomplished with each pass. Separation of the radionuclides to be transmuted from the spent fuel and from each other becomes more difficult and costly with each pass through the reactor because of the increase in higher mass isotopes and transuranics in the irradiated material.

[130] Bataille and Galley 1998, www.senat.fr/rap/o97-612/o97-061232 html
[131] Bataille and Galley 1998, www.senat.fr/rap/o97-612/o97-061232 html

Chapter III: Transmutation in critical reactors

The selective transmutation of long-lived radionuclides into stable or short-lived ones in critical reactors, although possible in theory, is, in practice, fraught with difficulties. These difficulties include:

- the fabrication of these radionuclides into fuel and targets requires additional shielding to protect the workers,
- the amount of any one radionuclide that, during a transmutation cycle, undergoes the desired transformation, that is, fission for the transuranics and transformation into a stable element for the fission products, is relatively small,
- the creation of higher mass transuranics which accumulate and pose new problems,
- the introduction of these higher mass TRUs radionuclides into the reactor affects the proper functioning of the reactor.

Transmutation of a large proportion of plutonium and higher actinides cannot be done in light water reactors alone, due to the inability of these reactors to deal with many non-fissile transuranic isotopes. Currently various combinations of light water reactors and fast reactors or sub-critical (ATW, Accelerator Transmutation of Waste) reactors have been proposed for waste transmutation schemes. Some transmutation schemes that would involve only sub-critical reactors have also been proposed. The next chapter will discuss schemes based on ATW reactors and the fuels that may be used in them. In this chapter, we will discuss the transmutation characteristics of light water reactors and fast reactors, as well as the special fuels and targets that may be used in these reactors.

Let us first examine the types of fuels that would be loaded onto these reactors, since this is essential to understanding the many practical technical difficulties with critical reactor transmutation options.

Types of fuels and targets

To transmute transuranic elements as well as long-lived fission products they must be fabricated into suitable forms for irradiation in reactors. There are broadly two ways in

which the radionuclides are fabricated for transmutation. The first consists of mixing the radionuclide homogeneously with the fuel; the second consists of putting the radionuclide in a specific target that is separate from the fuel. For example, to make MOX fuel (Mixed Oxide fuel, a mixture of plutonium oxide and uranium oxide), the plutonium is homogeneously mixed with depleted uranium. The exact composition of the fuel depends on the type of reactor. Of the large variety of fuel types, only the fabrication of plutonium in the form of MOX is currently done on an industrial scale.

The preferred fabrication for neptunium is its incorporation into MOX in a homogeneous manner. Americium, like neptunium, has been added to MOX but the preferred fabrication for this radionuclide is the heterogeneous way, in the form of targets. The fabrication into targets is also the preferred method for technetium-99 and iodine-129.

Table 7 shows fuel characteristics required for various reactors and the status of the technologies needed to fabricate these fuels.

Targets are the preferred forms for americium because the irradiation of americium in reactors results in the production of curium. We have also seen in chapter II that the separation of curium from americium is difficult, therefore there will be some amount of curium in the americium target before irradiation. Because curium-243, -244 are strong alpha and gamma emitters and curium-244 is also a significant neutron emitter (due to spontaneous fission), they pose serious handling, reprocessing, and fuel fabrication problems. If the americium is homogeneously mixed with the fuel, then the reprocessing of that fuel becomes more difficult due to the presence of curium. If americium is fabricated into targets it is proposed that, after irradiation, the targets would be stored for about hundred years to allow for the decay of curium-242, curium-243, and curium-244 into their corresponding plutonium isotopes.[132] The targets would then be reprocessed and the separated materials would be made part of the rest of the transmutation system.

Like most of the rest of the fuel fabrication systems proposed for transmutation, the fabrication of americium into targets is still at the research and development stage. The americium in the form of an oxide, nitride or carbide would be mixed with an inert-matrix. The inert matrix would be a ceramic (MgO for example) or a metal.[133]

If iodine-129 and technetium-99 are transmuted, they will most likely be fabricated into targets. The Experimental Feasibility of Targets for Transmutation (EFTTRA) group, a European collaboration, is carrying out the research for suitable materials. The fabrication of technetium into metal targets appears to be the method currently preferred by this group.

[132] Salvatores and Zaetta 1997, p. 111.
[133] OECD-NEA 1999b, p.145

Table 7: Status of fuel fabrication techniques for critical reactors

Types of reactor and fuel chemical form	Fuel type	Development scale	Irradiation scale
LWRs			
France			
Oxide fuel	Mixed oxide (MOX)[1]	Industrial	Commercial LWRs
	MOX-AmO_2	Computation model only	None
Fast reactors			
France			
Oxide fuel	MOX	Industrial/demonstration	Demonstration breeders
	MOX-NpO_2[2]	Experimental	Experimental
	MOX-AmO_2 [3]	Experimental	Experimental
	MOX-Minor actinide oxides [4]	Experimental	Experimental
Nitride fuel	UN-PuN[5]	Experimental	Experimental
Japan			
Oxide fuel	MOX-NpO_2[6]	Planning phase	None
	MOX-AmO_2[7]	Planning phase	None
	MOX-AmO_2-NpO_2[8]	Planning phase	None
Metal fuel	U-Pu-Zr-minor actinides-rare earths	Computation model only	None
Nitride fuel	U-Pu-N[9]	Experimental	Experimental
	Pu-minor actindes-[10]	Feasibility study	None
USA			
Metal fuel	U-Pu-Zr[11]	Experimental	Experimental

Sources:

Notes: [1] The fabrication of MOX in France is done by Cogéma in its Melox plant at Marcoule on an industrial basis. Belgonucléaire in Belgium also fabricates MOX for French reactors at its Dessel plant.

[2, 3, 4] These three fuels have been fabricated in Germany and irradiated in the fast breeder reactor Phénix (France).[134] Fuel rods were irradiated with high concentrations of americium and neptunium, as high as 20% for americium[135]

[5] Nitride pellet fuel pins have been fabricated in France for irradiation in Phénix, and in Germany for irradiation in the Petten reactor in Netherlands. The fabrication technology has been developed in Switzerland and India.[136]

[6, 7, 8] The irradiation, in JOYO, an experimental Japanese fast reactor, of MOX containing minor actinides is planned for around 2003.[137]

[9] Nitride pellets have been fabricated.[138]

[10] Dedicated burners would be loaded with high concentrations of minor actinides[139]

[11] More than a thousand fuel pins were fabricated and irradiated in the Experimental Breeder Reactor-II[140]

[134] OECD-NEA 1999b, pp. 142, 143.
[135] OECD-NEA 1999b, p.142
[136] OECD-NEA 1999b, p. 144.
[137] OECD-NEA 1999b, p. 143.
[138] OECD-NEA 1999b, p. 144.
[139]. OECD-NEA 1999b, p. 258.
[140] OECD-NEA 199b, p. 143.

Health and safety considerations relating to fuel fabrication of the transuranics

While the characteristics of these radionuclides present a health risk to the general public for the long term, they also present a health risk to the workers during their fabrication into fuel or targets. Low enriched uranium is mainly an alpha emitter and the precautionary steps taken during its fabrication involve worker protection from inhaling it, but its activity is considered low enough that glove boxes are not used. Almost all transuranics of concern are alpha emitters (with plutonium-241 being the exception). Their specific activity is far greater than low enriched uranium. All of them require handling in glove boxes. Moreover neptunium-237 and, in particular, its decay product protactinium-233 are strong gamma emitters. They require heavily shielded glove boxes during powder blending. Americium-241 and 243 are gamma emitters; americium-243 is also a source of neutrons. Its fabrication will require a high degree of automation. Curium, and in particular curium-243 and curium-244 are strong gamma emitters. Curium-244 is also a source of neutrons due to spontaneous fission.

Here are a few examples of the kind of facilities required for fuel fabrication:

- LEU fuel requires ventilated workplaces.
- The fabrication of plutonium into MOX fuel is conducted inside glove boxes,
- The addition of NpO_2 to MOX fuel requires the a 2mm thick lead shielding on the powder blending glove box to maintain the same external dose rates.[141]
- The gamma dose rate delivered by AmO_2 target pins is 2,780 times higher than for MOX fuel and it emits 7 times more neutrons per fuel pin. To compensate for this increase in radioactivity, a shielding of 4 cm of lead (for gamma protection) and 4 cm of resin (for neutron protection) is necessary.[142]
- The addition of curium to MOX fuel would require a polyethylene shielding, as thick as one meter, on the blending glove box to compensate for the increase of the neutron dose [143]
- The fabrication of americium-241 targets with 1 to 10 % lanthanides requires facilities which are fully shielded against gamma radiation and remotely operated[144]

As a consequence of the extra precautions needed to ensure worker protection during fuel fabrication for transmutation, the cost of MOX fuel fabrication will be significantly higher than the cost of fabrication of standard uranium oxide fuel. It is estimated that the fabrication of MOX fuel is four times more expensive than the fabrication of standard $UO_{2\ fuel}$ whose cost ranges from 275 to 300 dollars per kilogram of uranium.[145] It is estimated that the addition of actinides to the MOX fuel would the raise the cost of fabrication by another 20%.[146] It should be noted that this projected increase in cost is

[141] OECD-NEA 1999b, p.141.
[142] OECD-NEA 1999b, p. 146.
[143] OECD-NEA 1999b, p. 146.
[144] OECD-NEA 1999b, p. 47
[145] OECD-NEA 1999b, p.37.
[146] OECD-NEA 1999b, p.37.

based on very limited experimental work. Costs for large-scale nuclear projects such as breeder reactors have a way of escalating as they progress (see Chapter V).

Transmutation in LWRs

About two hundred years after the spent fuel is unloaded and the short-lived and medium-lived fission products have almost completely decayed away, plutonium contributes 90% of the radiotoxicity[147] of the spent fuel.[148] Therefore, if feasible, its nearly complete elimination from the spent fuel would theoretically result in a significant gain from the perspective of decreasing long-term risks. In reality things are more complicated because plutonium transmutation gives rise to new long-lived radioactive materials.

Transmutation as a waste management strategy cannot be implemented using LWRs alone. However, the LWR is the most common commercial reactor type in the world by far. Further, France as well as a few other countries (Germany, Switzerland and Belgium) are using commercial separated plutonium in the form of MOX fuel in many of their LWRs. Japan too has ambitious plans for this but, for a variety of reasons, these plans are stalled. The current use of MOX fuel, the established PUREX reprocessing industry, and the lack of economic competitiveness of MOX as a fuel relative to LEU, have led to proposals to view LWRs as the first segment of an elaborate transmutation system.

Transmutation of plutonium and other transuranic actinides in thermal reactors

For reactor safety reasons, only about 30 percent of a reactor core is loaded with MOX fuel rods, the rest being standard low-enriched uranium oxide fuel.[149] Currently, the plutonium content, that is all the plutonium isotopes, of French MOX fuel rods is 5.3%, but there is industry pressure to raise it to 8.65%[150]. Plutonium is both produced and consumed during reactor operation. In MOX fuel, plutonium supplies the fuel, which is fissioned and hence consumed, while the uranium-238 supplies the fertile material, which is converted into plutonium. On balance, there is a net reduction of plutonium in MOX fuel during light water reactor operation. By contrast, the low-enriched uranium portion of the fuel, which is 70 percent of the core, contains no plutonium at the time of fuel loading. During reactor operation, some of the uranium-238 in the LEU fuel is converted into plutonium. A part of this is fissioned in turn, but a part of it remains in the spent fuel.

During the operation of a typical reactor with 30 percent MOX core, the net consumption of plutonium in the MOX fuel itself is approximately offset by the production of

[147] For a definition of radiotoxicity see the glossary.
[148] Bataille and Galley 1998, http://www.senat.fr/rap/o97-612/o97-6123 html.
[149] Bataille and Galley 1998, http://www.senat.fr/rap/o97-612/o97- 61212 html. The enrichment in uranium-235 for LEU is 3.25%.
[150] The industry wants to increase the burn-up of the standard uranium oxide fuel. This would result in an increase of higher mass non fissile plutonium isotope compared to the fissile isotopes, in particular plutonium-239. An increase to 8.65% would be necessary to counter this negative effect. Bataille and Galley 1998. http://www.senat.fr/rap/o97-612/o97-61212.html.

plutonium in the LEU fuel. Typical details are as follows. A MOX fuel assembly contains 35 kg of plutonium before irradiation. After one cycle, 25 kg is left, that is there is a net plutonium reduction of 10 kg in each MOX fuel assembly. At the same time, there is production of 5 kg in a standard fuel assembly.[151] As inferred from above, for each MOX assembly, there are roughly 2 standard uranium fuel assemblies. Hence there is essentially the same amount of total plutonium in the spent fuel after irradiation as before, though the isotopic composition is degraded – that is, the plutonium in the spent fuel contains a larger proportion of higher plutonium isotopes (plutonium-240, -241, and 242) than fresh MOX fuel. The transmutation problem is also further aggravated by the fact that there is a substantial build-up of other transuranics like americium and curium in the MOX spent fuel, with the exception of neptunium. Table 8 shows the approximate initial and final minor transuranic element weights.

Table 8: Minor transuranic isotope growth in a light water reactor loaded with a 30% MOX core

Radionuclides	Initial loading kg/yr.	In spent fuel kg/yr.
MOX fuel (30% core)		
Minor actinides		
Neptunium-237	0	0.8
Americium (all isotopes)	0	23
Curium (all isotopes)	0	4.6
Sub-total, minor actinides	0	29
UO_2 fuel (70% core)		
Minor actinides		
Neptunium-237	0	6.2
Americium (all isotopes)	0	4.5
Curium (all isotopes)	0	0.39
Sub-total minor actinides	0	11
Totals for the reactor		
Minor actinides (all isotopes)	0	40

Source: based on Shapira, 1997 table I, p.11 and table II, p.17.

The reactor is a 900 MW LWR for both MOX and UO_2. The burn-up for the MOX fuel is 43.5 GWd/t. The burn-up for UO_2 fuel is 33 GWd/t. These figures are approximate as a result of possible mismatch between the burn-ups. They are given to illustrate the greater growth of minor actinides in MOX fuel. The inventories are taken 4 years after unloading for MOX and 3 years for UO_2

The radiotoxicity of spent LWR-MOX fuel after one cycle is about 8 times the radiotoxicity of spent LWR-UO_2 fuel. Of the total alpha activity, uranium and plutonium

[151] OECD-NEA 1999b, p.32

account for 30 %, the remaining 70% comes from Np, Am and Cm, with the last two being the main contributors.[152] Not shown in Table II is the production of higher isotopes of curium, some berkelium (which arises from the beta decay of curium) and californium (which arises from the beta decay of berkelium). Although the produced mass of these radionuclides is very small, the added radioactivity is significant. Moreover, it would increase with each pass of the fuel through the reactor. Further, californium-252 and some of the even-numbered curium isotopes undergo spontaneous fission, which becomes more probable with increasing mass number in the case of curium isotopes. Since spontaneous fission is accompanied by neutron emission, this added specific activity would require additional shielding for reprocessing and fuel fabrication, and therefore additional cost. It also complicates reactor operation.

Of the three minor actinides, neptunium, americium and curium, first americium and then, neptunium are the main contributors to the long-term radiotoxicity, while curium contributes relatively little to it.[153] While the transmutation of curium itself is not considered a priority, some of it will necessarily be transmuted along with americium, since separation of these two actinides poses serious problems, as we have noted.

We have chosen two examples to illustrate minor actinide transmutation in LWRs. The first is homogeneous recycling with MOX fuel – that is, the minor actinides are fabricated into the MOX fuel pellets. The second is heterogeneous recycling with standard uranium dioxide fuel – that is the minor actinides are fabricated into pellets that are placed into the pin separately from the fuel before being inserted inserted into the core.

The results of the computations for these two cases are shown in Table 9. They indicate that in the best case 70% of americium would be consumed. However, only 13 % would be fissioned, while 57% would be converted into higher mass transuranics. Much of the americium would be transmuted into curium, which, as we have noted, is a troublesome radionuclide. The rest, 30% is unchanged. The net result is that transmutation in this manner would, at best, eliminate 13% of the americium. Moreover, some of the fission products are long-lived. The prognosis for neptunium's best case is even worse – only 9% of it would be fissioned.[154]

[152] OECD-NEA 1999b, p. 49
[153] Boullis 1997, Figure 6, p. 83.
[154] The reactors vary from 900 to 1400 MW, the fuel is standard enriched uranium and MOX with various degrees of enrichments, there are many other parameters.

Table 9: Transmutation of neptunium and americium in LWRs

Radionuclide	MOX fuel Burn-up = 47.5 GWd/t Moderation ratio* = 3	UO2 fuel Burn-up = 42 GWd/t
Neptunium		
Consumption		
(kgd/TWh)	11	15
% fissioned	9	3
% transmuted	36	35
% left	55	62
Americium		
Consumption		
(kg/TWh)	10	8
% fissioned	6	13
% transmuted	36	57
% left	58	30

Source: Based on Salvatores and Zaetta, p. 109

*Moderation ratio is the ratio of the volume of the moderator to the volume of the fuel. A typical value for an LWR is 1.7. The moderation ratio assumed for MOX fuel is 3.0. The initial content of americium and neptunium would be 1% of the mass of heavy metal. The plutonium content of the MOX fuel would be 7.7%, in the case of Np transmutation, and 5.7 % for americium transmutation.[155]

The estimates of the low fission rates in Table 9 means that the use of light water reactors to transmute minor actinides would require a large number of passes through the reactor – entailing a correspondingly large number of reprocessing and fuel fabrication operations. Multiple passes create their own problems, however.

When neptunium-237 is irradiated with neutrons, there is a substantial production of plutonium-238. Plutonium-238 is a heat and neutron generator and its presence complicates separation and fuel fabrication for subsequent cycles. The proportion of plutonium-238 diminishes as the number of cycles increases but the simultaneous increase in the proportion of minor actinides becomes a serious problem.[156] The situation is generally similar with americium transmutation, which results in an increase in curium isotopes (see above).[157]

For these reasons, it has been proposed to store the irradiated americium targets in order to let the most problematic curium isotopes (curium-242, curium-243, and curium-244) decay into their plutonium isotopes (plutonium-238, plutonium-239, and plutonium-240). About one hundred years of storage would be required after which time it is proposed that the plutonium could be recycled.[158]

[155] OECD-NEA 1999b, p.151
[156] OECD-NEA 1995, p. 35
[157] Plutonium-238 results from neutron absorption by neptium-237, followed by beta decay. Curium isotopes are similarly created by beta decay of higher americium isotopes.
[158] Salvatores and Zaetta 1997, p.111

In conclusion, the transmutation of plutonium and the minor actinides in LWRs is very inefficient and it also increases the radiotoxicity of the spent fuel. In France, it has been proposed that plutonium in the form of MOX fuel be recycled only once in LWRs and then stored until a program to fission its long-lived transuranic components in fast reactors can be put in place.

Transmutation of fission products in thermal reactors

Two fission products have been studied for possible transmutation in LWRs: technetium-99 and iodine-129. Their neutron capture cross section for thermal neutrons is larger than for fast neutrons. However their transmutation in present day thermal reactors is difficult because large quantities need to be loaded in the reactors. This is because technetium is produced in reactors as a result of fission of U-235 in the fuel therefore, a large enough quantity of technetium must be loaded in target elements for transmutation in order to achieve a net reduction in technetium. Technetium and iodine require long irradiation periods. A calculation for the transmutation yield of technetium gives 11% per year, the calculation for iodine gives 3% per year.[159]

A reactor dedicated to the transmutation of these fission products would have to be loaded with a higher enriched fuel. This would be necessary in order to produce an excess number of neutrons to compensate for the neutrons absorbed by the technetium-99 and the iodine-129 and to create a high enough flux to achieve a satisfactory rate of their transmutation.

The transmutation of iodine-129 results in the production of xenon, which is a gas. This requires a venting of the target, which according to the Nuclear Energy Agency raises "considerable safety issues."[160]

Effects of the radionuclides on LWR reactor safety

The chain reactions in nuclear reactors must be properly controlled in order to prevent accidents. The chain reaction in a reactor is controlled by keeping close control of the number of neutrons and fissions that are occurring in it. When a constant amount of power is being produced, every fission in the reactor creates exactly one other fission, on average.[161] When a reactor is operating in this mode, its reactivity is said to be zero. When the power is to be increased, the reactivity is made positive (reactivity is "inserted" into the reactor) so that the number of fissions induced by each fission is slightly greater than one until the power reaches the desired level, at which time the reactivity is reduced to zero. When the power level is to be reduced or the reactor is to be shut, the reactivity is made negative – each fission induces less than one additional fission – until the desired condition of the reactor is achieved.

[159] NAS-NRC 1996, p.72
[160] OECD-NEA 1999b, p. 176.
[161] This applies only to critical reactors, the kind that are used today. For subcritical reactors, see Chapter IV

Changes in reactivity are achieved by controlling the number of neutrons and the spectrum of energy levels of the neutrons (since the number of fissions depends on both these parameters). Control rods made of neutron absorbing materials, most often boron, as well as the addition of neutron absorbing materials to coolants (in the case of some reactor designs) are used to control the neutron fluxes in reactors and hence the reactor power levels. Failure to control a reactor properly can result in severe accidents, of which the best known is the catastrophic explosion on April 26, 1986 of a reactor at Chernobyl in Ukraine.

Plutonium requires more control elements than a corresponding uranium reactor fuel due to its inherent nuclear characteristics.[162] Similarly, control requirements can change when too many fission products accumulate in a reactor, since these absorb neutrons, and tend to make reactor operation more difficult and less economical.

For these reasons, the addition of plutonium and minor actinides, which have different characteristics than uranium fuel, as well as the addition of fission products for transmutation into the reactor core, raises concerns regarding reactor safety as well as power distribution in the core of the reactor.[163]

MOX fuel use in LWRs makes the control of the reactor more difficult for the following reasons:

- The rate of fission of plutonium is higher than the rate of fission of uranium,
- The rate of fission of plutonium increases with temperature,
- The number of delayed neutrons per fission is less for plutonium-239 than for uranium-235 (0.2 percent versus 0.65 percent. The increase in the absorption of neutrons of intermediate energy (epithermal – that is faster than thermal neutrons but much slower than fast neutrons)[164] by Pu-239 and Pu-241 results in a decrease of the number of slow neutrons. As a result the control rods become less effective since they absorb mainly slow neutrons.

These phenomena increase the average energy reactivity of the reactor – that is, they shorten the response time for reactor control, for a given number of control elements. Hence, for the same reactor core, MOX fuel requires additional neutron absorbers in the form of control rods or the addition of boron in the cooling water,[165] they also limit the amount of plutonium which can be loaded in the reactor. For LWR MOX the amount of total plutonium (Pu-239 and other isotopes) is in the order of 5 % plutonium.

If the reactors are not designed to operate with plutonium fuel, or are not modified to increase the number of control elements to accommodate the use of plutonium fuel, this

[162] For a discussion of nuclear safety and reactors see Makhijani and Saleska 1999, chapter V. For MOX fuel use in light water reactors see SDA (Science for Democratic Action), Vol. 5 no 4 , February 1997
[163] NAS-NRC 1996, p. 53
[164] The energies of slow neutrons are a fraction of an electron volt, for the faster, epithermal neutrons they are up to one thousand electron volts, and for fast neutrons on the order of several thousand electron volts.
[165] Chemicals containing boron can be added for purposes of reactor control only in PWRs.

can increase the probability of severe accidents. In addition, since the reactors operating in a transmutation mode would generally have a larger inventory of transuranic actinides, including plutonium, the consequences of a reactor accident, should one occur, would be more severe.[166]

In addition to these reactor control and safety issues, the neutrons released from the fission of the plutonium have a higher average energy than the neutrons released from the fission of uranium. This increases the radiation damage to the reactor parts.

The transmutation of the minor actinides, neptunium and americium, also raises similar safety issues. These radionuclides can be put in the core of the reactor either mixed with the fuel (homogeneously) or separate from the fuel as targets (heterogeneously). In the latter case the targets may be placed in various ways, including at the periphery of the core. The introduction of these actinides affects the characteristics of the reactor in the following ways:[167]

- The reactivity of the reactor is affected initially: Because the cross sections of neptunium-237 and americium-241 are higher for capture than for fission with thermal as well as fast neutrons, the number of neutrons available for the maintenance of the chain reaction is reduced.[168] However, during the reactor cycle, this effect is mitigated by creation of more fissionable isotopes by neutron capture. These variations in reactivity (first a decrease and then increase) make issues of reactor control more complex. Moreover, since there is little experience with operating power reactors with large amounts of americium in them, the modeling of reactor operation and reactor safety is more difficult and less certain in transmutation modes.
- There is the possibility that the fuel temperature and number of fissions would simultaneously increase due to the addition of minor actinides to the core. [169]
- The presence of minor actinides would cause the reactivity to increase during loss of coolant incidents. Specifically, the higher neutron energy spectrum that results from the use of MOX fuel in the case of LWRs results in an increase in the rate of fission of neptunium and americium.

These problems, if not compensated for, can give rise to dangerous sudden increases in reactor power, which would increase the risk of severe accidents. These safety issues necessitate three kinds of considerations on the manner in which the transuranics would be loaded into a reactor:

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[166] Lyman 1999, pp. 7-10

[167] See OECD-NEA 1999, pp. 150-155 for a discussion of various safety and fuel placement issues related to minor actinide transmutation.

[168] This can be compensated by enriching the fuel in fissionable isotopes (uranium-235 or plutonium-239)

[169] In standard uranium fuel the increase of temperature in the fuel has a negative feedback, i.e., the effective cross section of uranium-238 for absorption increases and therefore less neutrons are available for fission and the reactivity for the reactor decreases. However with the addition of actinides, an increase of temperature results in an increase of the number of fissions and therefore a hardening of the neutron spectrum.

- The location of the transuranics in the reactor (in the core or on the periphery of the core): The impact on the characteristics of the core is the lowest when the minor actinides are placed on the periphery of the core and the most when placed in the core of the reactor.
- The form of the transuranics (homogeneous, heterogeneous or hybrid fuels): The transmutation of neptunium and americium is possible in homogeneous, heterogeneous, or hybrid modes. In the homogeneous mode the radionuclide(s) are placed in the core, in the heterogeneous mode there are generally placed on the periphery of the core. The hybrid mode is a combination of the homogeneous and the heterogeneous modes: one radionuclide is mixed with the fuel and placed in the core of the reactor and the other is fabricated into targets and put on the periphery of the core. The recycling of neptunium in the homogeneous mode is feasible, but the preferred mode for americium is heterogeneous
- The amounts of the transuranics loaded at one time: Their concentration in the fuel varies according to the type of critical reactor that is used, whether the mode of recycling is heterogeneous or homogeneous and whether they are placed in the core or on the periphery of the core. In general the concentrations are limited to 5% of the weight for transmutation in fast reactors and 1 to 2% in thermal reactors.[170]

In sum, the loading of transuranics into reactors as fuels to be transmuted will change the neutron economy of the reactor – that is the numbers and energies of the neutrons that are generated and how many fissions these neutrons trigger in the reactor core. There is some experience with the use of MOX fuel made from plutonium separated from LWR spent fuel in light water reactors, but there is very little experience on a large scale with the other elements that must be transmuted. The one possible exception is neptunium-237, which has been loaded into materials production reactors to make plutonium-238.[171]

Finally, as we have noted, the introduction of technetium-99 and iodine –129 into a light water reactor also changes its neutron economy, creating its own safety issues since fuel of higher enrichment would be required to compensate. There is also an additional safety issue of xenon gas venting related to the transmutation of iodine-129.

As a practical matter, the increase in the higher isotopes of plutonium (that is, pluotnium-240, -241, and –242), makes the repeated extraction and use of plutonium fuel in light water reactors impractical. Hence, for a large number of reasons, light water reactors cannot be effectively or efficiently used as transmutation machines to reduce the mass of transuranic radionuclides in spent fuel. In fact, their use increases the radiotoxicity of the spent fuel that must then be dealt with in reactors operating with fast neutrons. We will discuss critical fast reactors in this chapter and sub-critical reactors in the next.

Transmutation in fast reactors

The previous discussion highlights the severe limitations confronted by any program to transmute a large proportion of transuranic actinides in LWRs. Large numbers of passes

[170] Salvatores and Zaetta 1997, p. 108
[171] Pluonium-238 is used to make radioisotope thermoelectric generators (RTGs).

are needed, and even then there are substantial residual higher mass actinides that would continue to pose substantial problems. Further, the number of steps for recycling plutonium in LWRs is limited by the build up of higher plutonium isotopes, notably plutonium-240 and -242, which are not fissionable by slow (or thermal) neutrons. Fast neutrons have enough energy to fission even those nuclei such as plutonium-240 or plutonium-242 that cannot sustain a chain reaction with slow neutrons.

These problems have led to proposals for elaborate transmutation schemes in fast reactors, where fission using fast neutrons can, in theory provide more complete conversion of transuranic radionuclides into fission products. These schemes have their own serious limitations, not least of which is that fast reactors have not been successfully commercialized despite five decades of effort and enormous expenditures in many countries. Further, the problem of production of higher mass actinides will persist, even though it will be less pronounced than in the case of LWR transmutation schemes.

There are currently only three large fast reactors that are officially on the list of operating reactors. Of these, the 250 MW French reactor, called Phénix, is currently shut and is scheduled to be restarted in the year 2000 for use as an experimental reactor for transmutation of transuranic actinides. The other two large fast reactors, which are located in the former Soviet Union (one in Russia and one in Kazakhstan), are operating on uranium fuel with medium levels of enrichment.

Despite the poor record of commercializing fast reactors and the technical and economic problems that have caused major projects to be shut or abandoned in several countries, France and Japan are planning major programs to use fast reactors for waste transmutation. In France, high concentrations of plutonium up to 45% are envisaged, [172] despite the difficulty of addressing reactor safety issues involving fuel with a large proportion of plutonium.

Minor actinides would be mixed with MOX fuel for transmutation in fast reactors. One possible fuel composition that has been studied is 66% depleted uranium and 33% plutonium and minor actinides.[173] Table 10 shows that, although a larger fraction of neptunium and americium would be fissioned in the first pass through a fast reactor than in a light water reactor, the estimated rates are still relatively low. For neptunium, computer models estimate fission percentage from 24 to 27% in the first pass (compared to 3 and 9% in an LWR). For americium the corresponding range for a fast reactor would be 18 to 22% (compared to 6 and 13% for an LWR). Therefore, multiple passes will also be necessary for fast reactors. One study estimates that it would take as much as 225 years and as many as 15 passes through fast reactors to obtain a reduction of 88.4% in the mass of the mixture of transuranic actinides that is present in typical LWR MOX spent fuel. The study assumed that the fuel would be irradiated for 5 years and 12 years of cooling per cycle before reprocessing of the spent fuel[174]

[172] OECD-NEA 1999b, p. 156
[173] Fast reactors are designed to be loaded with MOX fuel containing a high percentage of plutonium, though no large-scale reactors have operated reliably for long periods with such fuel.
[174]OECD-NEA 1999b, p. 156

Table 10: Computer estimates of transmutation of neptunium and americium in an EFR (European Fast Reactor) type

Radionuclide	Homogenous fuel[1]	Heterogeneous fuel[2]
Neptunium		
Consumption (kg/TWh)	10	13
% fissioned	27	24
% transmuted by neutron absorption	33	36
% unchanged	40	40
Americium		
Consumption (kg/TWh)	9	14
% fissioned	18	22
% transmuted by neutron absorption	27	38
% unchanged	55	40

Source: based on Salvatores and Zaetta, p.109
[1] The neptunium and americium concentration in the fuel is 2.5%,
[2] The neptunium and americium concentration in the targets is 40%

Dedicated reactors to transmute minor actinides and fission products only are being studied in France and Japan. In this system, minor actinides separated from LWR or fast reactor spent fuel are fabricated into targets for irradiation in special reactors. The same would be done for technetium-99 and iodine-129.

The Japan Atomic Energy Institute is researching the use of two types of high neutron flux energy reactors: one lead-cooled, the other helium-cooled. The proposed chemical form of the fuel would be a nitride because of its good thermal properties, potential for allowing high burn-up and compatibility of the spent fuel with dry (electro-metallurgical) reprocessing. The amount of minor actinides fissioned in these reactors would be of the order of 250 kilograms per year per pass through the reactor. Multiple passes would be required.[175]

In France the prevailing opinion within the nuclear reactor establishment is that only fast reactors should be used for the transmutation of fission products. Although the cross section of fission products in fast reactors is smaller than in LWRs, the high neutron flux in a fast reactor provides certain advantages. First the actinides would be transmuted by fast neutrons that would then be slowed down for the transmutation of fission products. (This suggested approach relies on the relatively small amount of moderation that does occur due to collisions between neutrons and sodium nuclei in the reactor core.)

Computations for the transmutation of technetium-99 targets placed in the core of a fast reactor show that about 166 kg of technetium-99 can be transmuted per year in a 1,500

[175] OECD-NEA 1999b, p. 257.

megawatt fast reactor. This amount of Tc-99 is produced each year in about five to six LWRs of 1,000 megawatts each. The effective transmutation half-life is about 26 years. In the case of iodine-129, computations show that 22 kg can be transmutation with a transmutation half-life of 44 years.[176]

Fast reactor safety would be a major issue in case of the use of such reactors as waste transmutation machines. The largest amount of experience with fast reactors in the world by far is with the liquid sodium-cooled fast reactor. Tens of billions of dollars have been spent of developing these reactors, including several large reactors with capacities of more than 100 megawatts electrical (most commercial light water reactors have capacities around 1,000 megawatts electrical). Technical operating problems have been rife, and there have been several major accidents. The very first fast reactor, Experimental Breeder Reactor 1 built in Idaho, suffered a partial meltdown in 1955. The most recent accident was a major leak, in 1995. of liquid sodium and subsequent fire that occurred in the Japanese Monju reactor, not long after it had been commissioned. It remains shut. The largest breeder reactor in the world, Superphénix, built in France, was prematurely shut in 1998 due to persistent operating problems. Besides meltdown accidents, that can occur in light water reactors also, sodium-cooled fast reactors can suffer from sodium leaks and fires, failures of cooling equipment handling liquid sodium, and potential catastrophic accidental super-criticality accidents. These well-known concerns with fast reactor safety would be complicated further by the introduction of minor actinides as well as fission products in the form or target rods. It is somewhat mysterious how so many plans for the use of fast reactors with exotic fuel and target core configurations are being made, when even the operation of these reactors with the fuel for which they were designed, MOX fuel with about 30 or 40 per cent plutonium content, has not been successful on a routine, reliable basis.

In addition to these general concerns about sodium-cooled fast reactors as a viable technology, there are a whole host of issues associated with their use as waste transmutation devices. Broadly speaking, many of these issues are similar to the ones that we have already discussed in the case of light water reactors, with the exception of the class of issues related specifically to the exact composition of the thermal neutron energy spectrum. This issue does not arise in fast reactors since they do not operate on thermal neutrons. The question of changes in reactivity, control of the reactor, the probabilities and consequences of accidents, would all tend to raise risks. Further, relative to LWRs, there is very little operating experience with fast reactors. The experience using plutonium fuel is even more limited. There is only one large reactor, the French Phénix, where experiments suitable for determining safety and performance issues are scheduled to be carried out on a significant scale and this reactor is due to be shut in 2004. Only seven cycles of experiments are planned in this reactor between the year 2000 and its planned permanent shut down.[177]

[176] OECD-NEA 1995, p. 26
[177] Bataille and Galley 1998 at www.senat.fr/rap/o97-612/o97-61234.html

The development of fast reactors as reliable devices in normal operation and their further development for use as waste transmutation devices poses severe hurdles for a critical reactor based transmutation system

Chapter IV: Transmutation in Accelerators

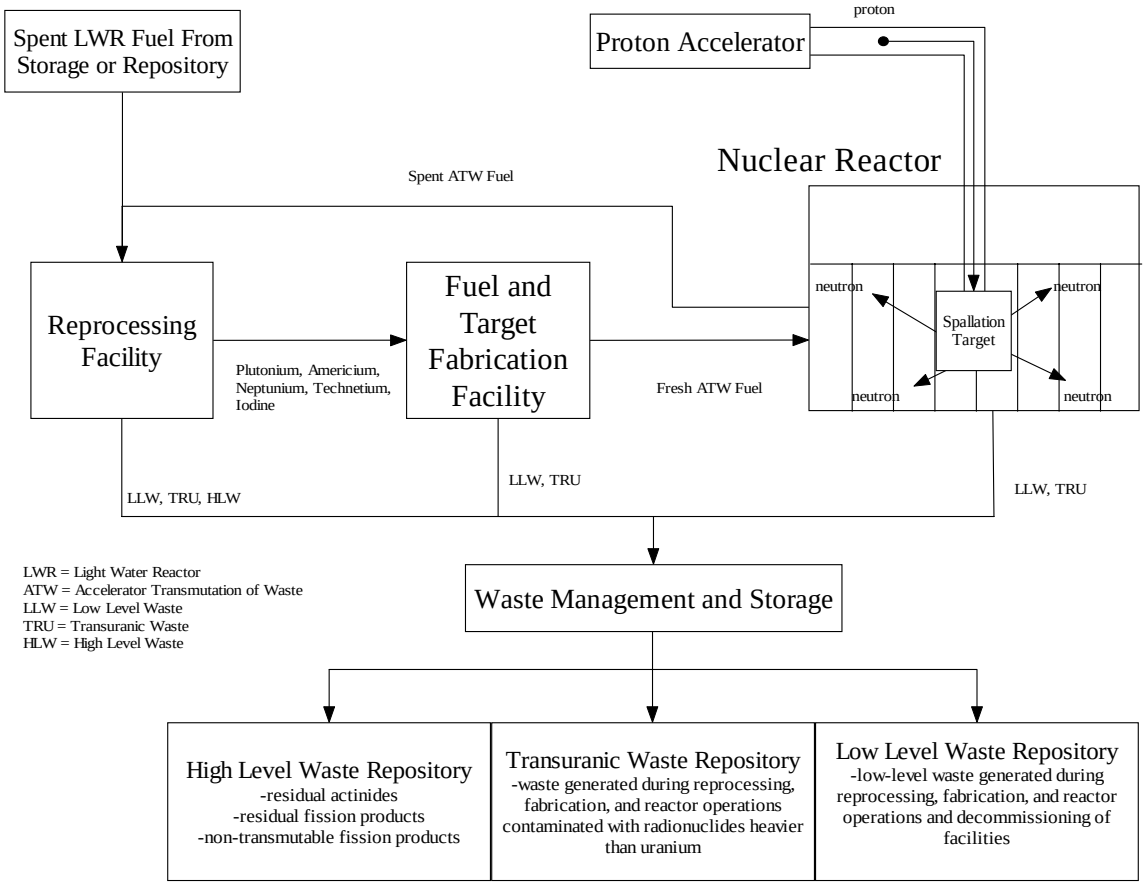
The other major category of transmutation system (in addition to critical reactors) is accelerator-based systems. There a number of proposals worldwide for Accelerator Transmutation of Waste (ATW).[178] However, all of them operate on the same basic principles and include the same basic components:

1. An accelerator of protons
2. A spallation target (for neutron production)
3. A sub-critical reactor fueled with plutonium and minor actinides and, in some cases, fission products and/or fertile fuel (fuel which produces more fissionable radionuclides).
4. A front-end and back-end chemical processing capability for preparing current spent fuel and/or liquid high level waste and spent ATW fuel to pass through the reactor.

The differences between them are in the choices of neutron spectrum, type of accelerator, composition of the fuel (including whether or not fission products will be transmuted and whether fertile fuel will be used), and choice of reprocessing technology. This chapter will provide an overview of the different components of accelerator-based transmutation systems and some examples of the leading programs for ATW. A typical transmutation system is depicted in Figure 14.

[178] A number of acronyms are used to describe ATW, however, we have tried to maintain consistency with the use of ATW. Other acronyms used at various times are ADS (Accelerator Driven Systems, which refers to any accelerator and sub-critical reactor system, whether or not it is used for waste transmutation) and ADTT (Accelerator Driven Transmutation Technology).

Figure 14: Accelerator Transmutation System



It is important to note that, despite their name, ATW systems do not use accelerators for the actual transmutation process. Transmutation reactions occur in the sub-critical reactor, not in the accelerator. The accelerator's function is to produce supplemental neutrons for the reactor. Thus, ATW systems are in fact a type of nuclear reactor. As such, it is also possible to use the energy released in the nuclear fission process to generate electricity for commercial sale. This is used to offset some of the large costs of these systems (see Chapter V).

There have been a large variety of ATW systems proposed by a number of groups. While all of them share some common characteristics, they also differ widely on key components such as fuel type, coolant, and accelerator type. In order to provide concrete and consistent information this chapter will use the U.S. ATW program as a leading example of a linear accelerator based program and the Energy Amplifier as a leading example of a cyclotron based program. These two appear to be the most advanced and developed proposals as well as having the greatest amount of publicly available information. However, ATW programs in other countries are very similar in many respects and, thus, the information in this chapter is widely applicable. This chapter will not cover transmutation proposals based on electron accelerators or photo-transmutation, as they are not an active area of inquiry. However, even with these systems, much of the technology (e.g. for separation of radionuclides) would be similar.

Furthermore, through a series of international conferences and meetings there appears to be a growing consensus on the parameters for an ATW system based on linear accelerators and with fuel composition, separations technologies, and coolant/spallation targets similar to the US system described. For this reason, linear accelerators systems will be given more attention. Again, however, there are definite similarities in all of the proposals. A team based at the European Organization for Nuclear Research (CERN) has developed a concept using cyclotrons rather than linear accelerators.[179] The team is led by Dr. Carlo Rubbia, a former director of CERN, and is called both the Energy Amplifier and the Rubbiatron. At this time it appears to be the only one based on a cyclotron, but much of the rest of the technology is the same or similar to the US and other systems.

It should also be noted that this field is a rapidly changing one. Even the Los Alamos National Laboratory (LANL) proposed ATW program has evolved rapidly over the past few years from a molten salt thermal system to a solid-fueled fast neutron system with liquid lead-bismuth as both coolant and spallation target. Therefore, this chapter does attempt to remain as broad as possible and then provide some more specific examples at the end.

Accelerators

There are two major types of accelerators being considered for transmutation: linear accelerators and cyclotrons. They are described below.

Linear Accelerators

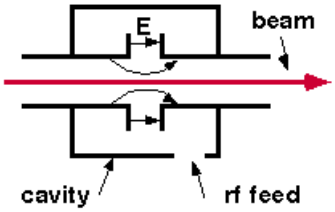
As the name indicates, linear accelerators (or linacs) increase the energy of the proton over a straight path. Ion accelerators start with an injector system which produces the ions, extracts them, and prepares them for acceleration in the linac. The ion source is a plasma of hydrogen ions created by heating the gas (for example, a microwave power source and a magnetic field could be used to create a plasma discharge). A voltage difference is used to extract the ions and a low-energy beam transport device focuses the ion beam and prepares it for acceleration.[180]

Linacs use electric fields to accelerate the charged particles. The electric field is created through the application of an alternating current radio-frequency (RF) source. The linac consists of a series of tube electrodes connected to the RF source (a simple diagram of an RF cell is shown in Figure 15). The electric field occurs in the gap between successive tube electrodes. The alternating current of the source means that the electrodes continuously switch back and forth from positive to negative. Since successive electrodes have opposite charges, an electric field is created in the gaps. The accelerator is designed so that ions pass through the electrode gaps at exactly the correct times so as to be accelerated by the electric field in pulses which are coordinated to occur

[179] The primary mission of CERN is to conduct basic nuclear physics research, concentrating mainly on basic particle physics. CERN is a joint venture among a number of governments. The research relies greatly on the use of particle accelerators. While the initial work on the Energy Amplifier was done at CERN, it appears that any further development would be done outside of the laboratory.
[180] Krane 1988, pp. 560-561, 588-593 and DOE SRS 1997, p. A-3.

when the field lines are in the direction of beam travel.[181] While RF accelerators are a well-established technology, they face certain limitations. Aside from expense, the RF accelerator by definition is tied to the frequency of its RF generator, which can be doubled or tripled, but is essentially limited. Since the beam current is proportional to the frequency, this imposes limitations on the beam current.[182] In the context of transmutation, this in turn imposes certain limitations on the potential neutron production of an accelerator-target system.

Figure 15: Simple RF Cell



Source: Lawrence Berkeley National Laboratory Website

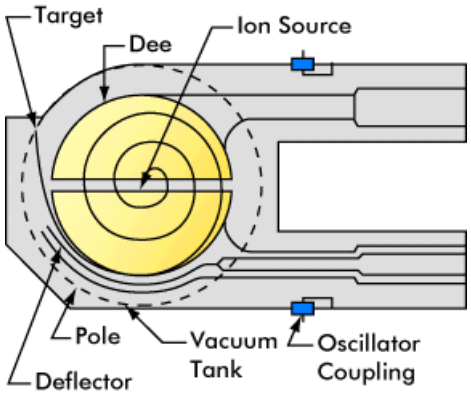
This is a highly simplified explanation of how a linear accelerator operates. There are different types of accelerating cavities that can be used in conjunction with each other, and the proposed ATW linac does use multiple types of tubes. However, the details of the accelerator are not important for understanding ATW.[183]

Cyclotrons

Ion creation and introduction into the accelerating structure is essentially the same for cyclotrons as it is for linacs. It is the acceleration of the protons that differs greatly. Cyclotrons use magnetic fields to accelerate the proton in a large ring (see Figure 16). When the proton has reached the desired energy level it is redirected at the target. The cyclotron itself consists of two large semi-circular chambers, magnets, and a source of alternating voltage. The voltage source creates electric fields in the gap between the two semi-circles. The magnetic field bends the proton beam in a circular path. The result is that every time the protons pass across the gap, they get a small "kick" in energy. When they are in the chambers they are not subject to the electric field lines and travel in a semi-circle. The protons spiral out from the center of the chambers gaining energy and speed.[184] As with the description of the linacs, this is a simplified explanation. The Energy Amplifier design would use three successively larger cyclotrons in series to accelerate the protons. Each cyclotron has multiple gaps rather than the basic single gap device described above, giving them a pinwheel look.

[181] Krane 1988, p. 588-589.
[182] LBL HIF website.
[183] Readers interested in learning more about linacs should consult Krane 1988, DOE 1999d, Wangler 1998, Lee 1999,and the ATW Roadmap 1999b.
[184] Krane 1988, p. 571

Figure 16: Cyclotron



Source: Stanford Linear Accelerator Center

Cyclotrons have the advantage of being more compact than linear accelerators. The two large cyclotrons proposed for the Energy Amplifier have external diameters of 10.5 m and 16 m. By comparison, the linac proposed for ATW in the Roadmap report is 296 m long.[185] However, cyclotrons are also limited in the currents they can achieve (and therefore the power of the accelerator for a given proton energy).

Current state of accelerators and levels necessary for transmutation

The ATW project in the United States builds upon advances anticipated through two other major linear accelerator projects. The first is the Accelerator Production of Tritium project. This program, chosen as the backup for production of tritium in a commercial reactor, would use accelerated protons to generate neutrons through spallation. Those neutrons would then strike lithium targets to produce tritium for the United States nuclear weapons arsenal.[186] The second project is the Spallation Neutron Source (SNS), a multi-billion dollar science program to develop a neutron source for a variety of scientific experiments. The Oak Ridge National Laboratory is the preferred site to construct the SNS.[187]

While there are differences between the two projects (e.g. the APT would be a continuous proton beam and would use a tungsten target while the SNS would be operated in pulse mode and use a mercury target), the basic accelerator designs are the same. The proposed accelerators are quite large (protons would be accelerated to approximately 1 GeV with a beam current of 100 mA resulting in a power of 100 MW).[188] The accelerators proposed for ATW would have the same proton energy, but would have a lower current (on the order of 45 mA resulting in a beam power of 45 MW).[189] While other programs may have slightly different accelerator designs, the basic parameters will

[185] Rubbia et al 1997a, p. 231 and ATW Roadmap 1999b, p. 5
[186] See Zerriffi 1996 for a discussion of the role of tritium in nuclear weapons.
[187] DOE 1999d, p. S-1
[188] GeV – giga-electron-volt. This is a measure of the energy of the proton. MA – milli-amperes. This is a measure of the accelerator's current (the amount of electricity per second being conducted by the accelerator, essentially it measures the number of protons that are being accelerated). MW – megawatts. This is a measure of the total power and is the multiplication of the current of the accelerator and the energy of the protons.
[189] ATW Roadmap 1999b, p. 5

be similar. This is a result of the desire to have reactors of a certain power and from the physics of the spallation process (discussed below).

In both the Accelerator Production of Tritium (APT) case and the Spallation Neutron Source (SNS) case, the accelerators are significant advances from the current state of the art in linear accelerators. These advances would build upon the LANSCE accelerator at Los Alamos National Laboratory, which is an 800 MeV proton accelerator that operates at 17 mA in a pulse mode.[190] The development program to increase the power and, in the case of APT, to switch from pulse to continuous wave mode factor into the large costs associated with these programs. The design and construction costs for the SNS are estimated at approximately $1.22 billion with an additional $220 million in indirect costs (e.g. necessary R&D).[191] Note, however, that this is for the entire facility, not just the accelerator and spallation target sections (i.e. it includes the beam lines for neutron experiments and associated buildings, etc.). The APT, which was designated the back-up technology for tritium production, was estimated in 1995 to have a project cost of $2.5-3 billion and a life cycle cost (including operations, maintenance, decontamination, and decommissioning) of $9.1-12.4 billion.[192] However, like all major projects, those involving accelerators can experience substantial delays and cost overruns. For example, the Dual Axis Radiographic Hydrotest Facility at Los Alamos, which uses electron accelerators, went from an estimate of $30 million in 1988 to an estimate of around $270 million in the most recent budget request.[193] A more detailed discussion of cost issues is presented in Chapter V.

The cyclotron-based systems would also require advances in accelerator technology beyond the current state of the art. According to Dr. Rubbia's team at CERN, the cyclotron required for the Rubbiatron or Energy Amplifier would have to produce a current of protons about ten times higher than present cyclotrons.[194] Currently, the highest power cyclotron accelerates protons to 590 MeV with a current of 1.6 mA.[195] While the power of cyclotrons can be increased, there is doubt that they could reach power levels above 10 MW.[196] For both linear accelerators and cyclotrons, these advances are considered significant, but not the major obstacle to implementing transmutation systems[197].

One area in which both linacs and cyclotrons will need to improve dramatically, however, is in the reliability of the beam. Current accelerators suffer from frequent interruptions of the beam, called beam trips. Accelerated beams composed of charged particles are subject to a variety of instabilities. The beams are monitored and if a

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[190] ATW Roadmap 1999b, p. 3. This accelerator operates with a 6% duty cycle (meaning that rather than having a continuous beam, it has pulses and that the accelerator is actually accelerating protons 6% of the time).
[191] Fiscal Year 2001 Budget Request of the Department of Energy, Science, Basic Sciences, Project 99-E-334 Spallation Neutron Source.
[192] DOE 1995, pp. 6-8 and 6-9.
[193] Fiscal Year 2001 Budget Request of the Department of Energy, weapons Activities, Construction, Project 97-D-102 Dual-Axis Radiographic Hydrotest Facility.
[194] Rubbia et al. 1997a, p. 230
[195] ATW Roadmap 1999b, p. 3
[196] ATW Roadmap 1999b, p. 3
[197] ATW Roadmap 1999e, pp. B-1, B-2

problem is detected in the beam, the beam is shut down. This is called a beam trip.[198] In most cases these beam trips last for only a short period of time (less than a minute). However, even these short beam trips can have a negative effect on the performance of the transmutation system. First, it can cause power fluctuations or transients in the reactor which could be a safety problem and which could damage fuel elements. Second, fluctuations in beam power can cause thermal stresses as the neutron source is turned on and off and the heat levels in the reactor fluctuate. This can cause accelerated degradation of key materials in the reactor. Third, beam trips would cause interruptions in the flow of electricity to the electrical grid, which would be a problem for reliability of the grid (the loss and then resumption of 3000 MWe could cause problems for grid stability). Fourth, it would effect the economic operation of the reactor. Longer beam trips (greater than 10 minutes) would actually necessitate a long re-start of the power plant (an hour or more) during which time transmutation would not occur and electricity would not be produced. Repeated beam interruptions of this duration would greatly reduce the capacity factor of the facility. As a result development of high reliability components will be necessary to reduce the beam trips from the current level of approximately one per hour to a few per year.[199]

The ATW Roadmapping report favors linacs over cyclotrons due to the high power levels they can achieve. A higher power accelerator driving multiple sub-critical reactor cores is considered to have advantages in terms of capital and operating costs over a low-power accelerator driving a single core.[200]

Separations

Depending upon the technology choices made (e.g. neutron spectrum, fuel type, etc.) a wide variety of separation technologies could be used for accelerator transmutation of waste. Programs based upon oxide fuels would likely continue to use some form of PUREX processing. Meanwhile, programs based on metal fuels (such as the current plans for ATW developed by LANL) would use some form of electro-chemical processing. Others may use a combination of technologies. For example, while pyro-processing is used for separation of actinides and fission products in the US ATW scheme, the incoming LWR fuel would actually undergo a PUREX-like process first in order to separate the uranium (this process is called UREX). Descriptions of these technologies can be found in Chapter II.

Overview of Spallation

The ATW concept requires a supplemental source of neutrons. In the critical reactor concepts discussed above, that neutrons all result from fission of fuel in the reactor. The accelerator based systems have an added source of neutrons from spallation (while ATW systems do not have critical configuration, there would still be a significant number of fissions and neutrons from fission reactions). The word spallation comes from the word "spall," which means to "chip off." Spallation is a nuclear reaction in which an

[198] For more information on accelerators and beam instabilities see Wangler 1998 or Lee 1999.
[199] See IAEA 1997a, p. 81, ATW Roadmap 1999b, p. 13 and pp. 28-29, Takizuka et al 1998b, p. 390
[200] ATW Roadmap 1999b, p. 4

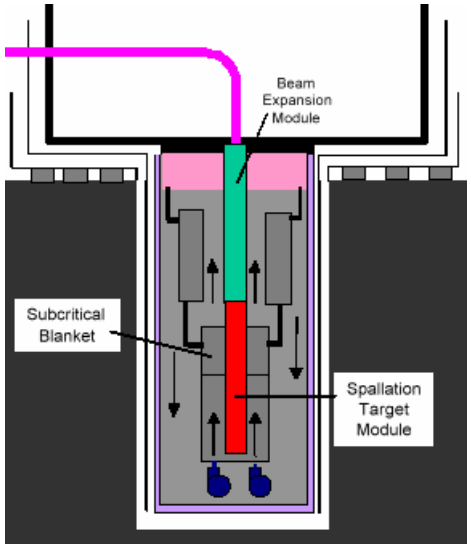
incident particle (the particle that hits the target, in this case a proton) is of high enough energy that the target nucleus does not form a compound nucleus. Instead, a variety of direct reactions can take place. First, secondary particles such as pions, neutrons, and protons can be ejected at a lower energy, but in the same general forward direction of the incident particle. Second, the excess energy of the target nucleus is removed by the "evaporation" of nucleons, mainly neutrons but also alpha particles, leaving behind a "spallation product". These two reactions give the reaction its name, spallation, because the particles are essentially chipped off of the original nucleus. Figure 18 provides an illustration of the spallation process. The target nucleus can also undergo high-energy fission resulting in fission products and neutrons. In addition the secondary particles can go on to induce other spallation reactions if the target is thick enough.[201]

As a result of the spallation process neutrons are produced which then enter the transmutation reactor. There are two sets of neutrons produced. The first are emitted in a forward direction as secondary particles from spallation. The second set of neutrons is emitted from the "evaporation" of excess energy in the target nuclei and from fission. These neutrons are emitted isotropically, that is, in all directions, as indicated in the figure. The transmutation fuel can therefore be positioned surrounding the spallation source as indicated in Figure 17.[202]

In the case of an ATW system the target is thick enough and the incident protons energetic enough to have a cascade of spallation reactions. The majority of the produced neutrons (~90%) are produced through evaporation and are emitted isotropically (i.e. in all directions).[203] The rest are emitted as secondary products and are thus primarily in the direction of the incident proton. However, the large proportion of isotropic neutrons allows for the fuel to surround the spallation region. The length of the spallation region and the energy of the proton are related. A higher energy proton will cause a larger number of cascading reactions and will thus require a longer spallation target region. There is also an optimal energy region for neutron production. It appears that ATW proponents are converging on a parameter of approximately 1 GeV for the incident protons which requires a spallation target approximately a meter thick.[204] For a lead-bismuth (LBE) target and proton energy of 1 GeV, then approximately 30 neutrons are produced for every incident proton that hits the target.[205]

[201] Gudowski 1997, p. 5-6. and Bowman 1998, p. 510-511
[202] Gudowski 1997, p. 5-6. and Bowman 1998, p. 510-511
[203] Bowman 1998, pp. 510-511. It should be noted that this means some of the neutrons travel backwards along the line of the proton beam. This contributes to the radiation damage experienced by the beam window (which separates the accelerator tube, which is under vacuum, from the spallation target) as well as results in neutron activation within the accelerator structure.
[204] Bowman 1998, pp. 510-511.
[205] ATW Roadmap 1999f, p. 15.

Figure 17: Spallation Source and Surrounding Fuel

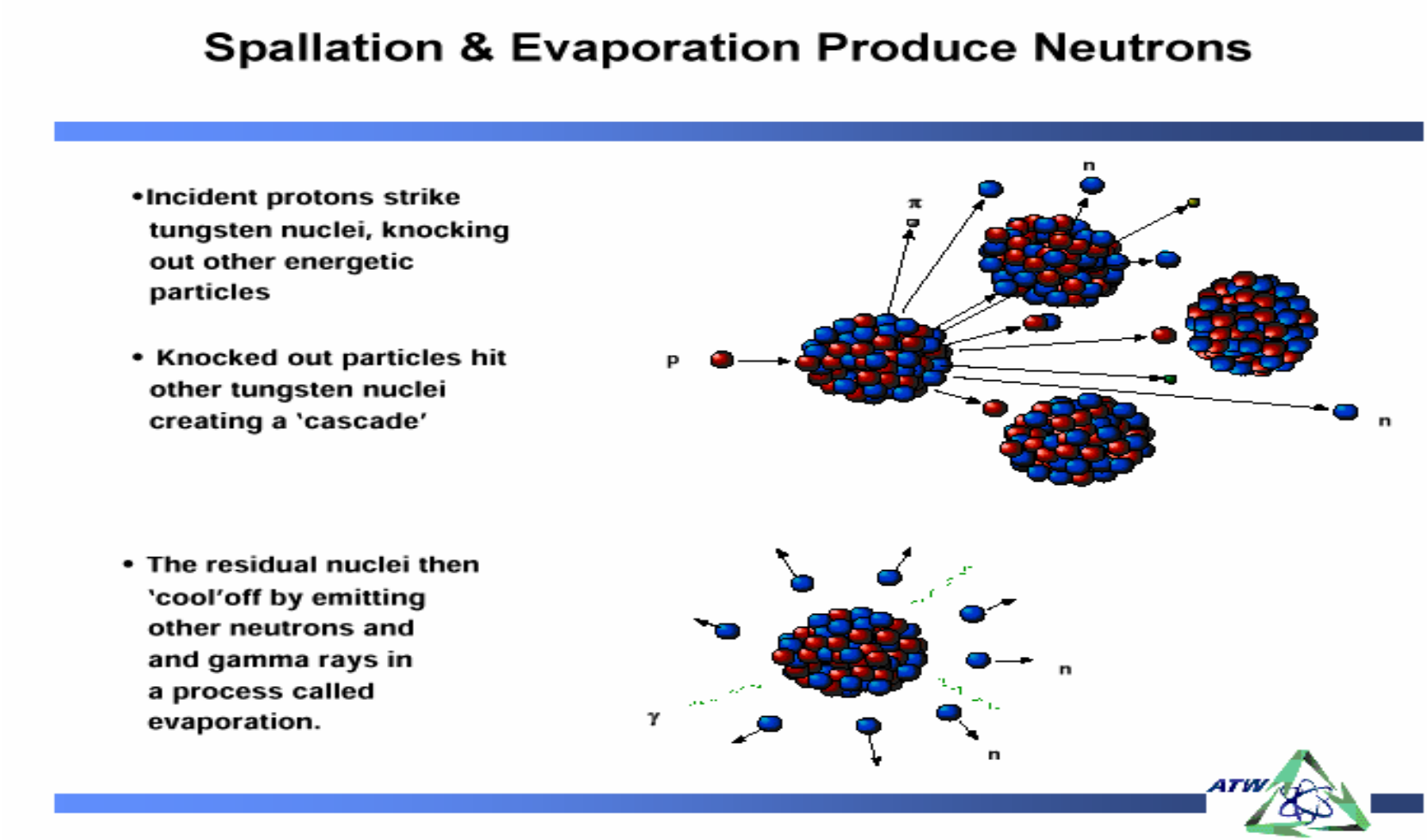


Source: ATW Roadmap 1999e, p. 4.8

The average number of neutrons produced by spallation per incident proton does not significantly vary for a given proton energy (e.g. 1 GeV).[206] However, more neutrons can be produced by increasing the current of the accelerator (i.e. accelerating more protons of that particular energy). The current is a measurement of the amount of electricity being conducted per unit of time and is measured in amperes (or more commonly milliamperes or one-thousandths of an ampere). The accelerators proposed in the ATW Roadmap would be 45 milliamperes. The power of the sub-critical reactor is therefore partially determined by the current of the accelerator and partly determined by the energy of the particles being accelerated. In the above case, an accelerator accelerating protons to 1 GeV with a current of 45 mA would have a power of 45 megawatts (MW). The power can, therefore, be adjusted by adjusting the accelerator current. This would be used to compensate for changes in the reactivity of sub-critical reactors as the fuel is consumed or the reactivity of the sub-critical core changes.

[206] However, the number of neutrons produced does depend on other factors, such as the spallation target material and configuration.

Figure 18: The Spallation Processes



Source: Gregory J. Van Tuyle, Los Alamos National Laboratory, "Options for the ATW Transmuter," Presentation to the Nevada ATW Workshop, May 8, 2000

Types of Fuels and Coolants

As discussed in Chapter I and Appendix C, there are a variety of different types of fuels and coolants possible for nuclear reactors (both critical and sub-critical). Both solid and liquid fuels have been proposed for use in sub-critical reactors. Coolant choices include sodium, lead, lead-bismuth eutectic, and gas. Lead-bismuth eutectic will discussed in this section as it has received significant attention lately as a candidate coolant despite the limited experience in using this material as a coolant.

Solid vs. liquid fuels

Accelerator-based systems have been proposed with both solid fuels and liquid fuels. Solid fuels would most closely resemble nuclear reactor fuels currently in use or which have been developed for breeder reactor programs. These fuels could be ceramic oxides or metal fuels. Liquid fuels have never been used commercially but have been part of nuclear research programs and experimental reactors. Each fuel poses certain advantages and disadvantages for proponents of transmutation.

Solid fuels are made using a matrix that contains a certain percentage of fissionable fuel. The matrix can be classified as either inert or fertile. Inert matrices are made of a material that does not produce fissile materials when irradiated. Zirconium, proposed for use as the matrix in the US program, is an inert matrix. Neutron irradiation of zirconium produces radioactive activation products (including one long-lived activation product), but not a radionuclide that can undergo fission.

Fertile matrices are made of a radionuclide that absorbs neutrons and results in a radionuclide that can be fissioned (usually after some intermediate radioactive decays). These include thorium-232 (which results in the production of fissile U-233) and uranium-238 (which results in the production of Pu-239). Fertile matrices have been proposed as part of accelerator transmutation, either initially or to produce more fuel once existing stocks of conventional reactor fuel have been processed. In that way, ATW has been closely linked to the continuation of nuclear power indefinitely (particularly through the Thorium-Uranium cycle). This is discussed in more detail in Chapter V. The enrichment of fissionable material in the fuel can vary. The current US plan calls for 25% actinides and 75% zirconium.[207] With a solid fuel matrix, the long-lived fission products would be fabricated into separate transmutation targets.

Liquid fuels have a carrier material that performs the same function as the matrix in the solid fuel. The fuel usually proposed is a molten salt. The molten salt would consist of a carrier material (such as lithium fluoride) and the materials to be transmuted (such as Pu fluoride).[208] Another liquid fuel that has been proposed is a heavy water slurry in which the target radionuclides would be dissolved in the liquid heavy water (for example as suspended plutonium oxide particles).[209] While more experimental than solid fuels, the liquid fuels were supposed to provide the benefit of continuous on-line

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[207] ATW Roadmap 1999f, p. 21

[208] NAS-NRC 1996, pp. 278-279

[209] NAS-NRC, pp. 276-278. Heavy water has deuterium atoms rather than hydrogen atoms (D_2O rather than H_2O). Deuterium is an isotope of hydrogen with one proton and one neutron.

reprocessing. However, it appears that the difficulties posed by liquid fuels may be too daunting. Historically, molten salt reactors have had significant operating difficulties that have prevented their theoretical advantages from being realized in practice. Work on molten salt reactors ended by the seventies.[210] The US proposal has recently switched to a solid fuel. Another disadvantage of liquid fuels is that they contain two fewer levels of containment. The solid fuel itself acts as a containment matrix for most fission products. Solid fuels also often have a layer of cladding material surrounding them. This cladding material can contain the gaseous fission products that are produced.

Coolants

Coolants must move the high heat levels caused by fission and radioactive decay away from the fuel and to heat exchangers. Steam is produced in the heat exchangers, which drives a turbine generator to produce electricity. Coolants can be either gas or liquid. However in the case of ATW, most proposals are based on liquid coolants.[211] Lead-Bismuth Eutectic (LBE) as both coolant and spallation source has emerged as a front-runner, particularly in the United States and Russia. An eutectic is a mixture of two materials with the lowest possible common melting point for the combination.[212] The LBE coolant was originally implemented by the Soviet military for cooling on their nuclear-powered submarines (the United States also experimented with lead coolants early on, but abandoned the program).[213] As a coolant, LBE has certain advantages[214]:

- The lead and bismuth have high atomic numbers and therefore will not moderate the neutrons. The fast neutron spectrum results in more effective fissioning of actinides and higher neutron production.
- LBE is not reactive like liquid sodium metal, the coolant used in many breeder reactor research programs using fast neutrons. This reduces operational problems that have plagued breeder programs and also reduces worker and public health risks.
- LBE has a low melting point (123.5 ^{o}C) but a high boiling point (1670 ^{o}C) allowing for a large temperature range of operation, particularly in abnormal conditions.
- The lead can also act as the spallation target, eliminating the need for a different spallation target material.

[210] NAS 1995, p. 190
[211] At the recent ADTTA '99 conference in the Czech Republic, two French officials from Framatome and the CEA presented a proposal for a gas-cooled accelerator based system. Their reasons for investigating the feasibility of gas-cooled plants is the greater operating experience with gas cooled reactors and greater ease of maintenance in comparison to liquid-metal cooled reactors. (Carluec 1999, p. 5)
[212] ATW Roadmap 1999e, p. 3-4
[213] ATW Roadmap 1999f, p. 15-21
[214] Venneri et al. 1998, p. 7

However, LBE also comes with some risks and uncertainties that would have to be resolved:

- Lack of experience outside of Russia
- Lack of experience with LBE as spallation target
- Corrosive effects of LBE
- Health hazards of lead and bismuth
- Build-up of neutron activation and spallation products, particularly Po-210 (138 days) which can pose operational concerns, long-lived Pb-205 ($1.5x10^7$ years) and the volatile Kr-85 (10.72 yr.), Xe-127 (36.4 days) and Hg-194 (520 years). The production of spallation products and neutron activation of the target causes concern both for operations and for disposal of the lead during decommissioning.[215]

For any target-coolant combination, there are advantages to having a separate loop for the reactor coolant. This avoids having spallation products circulating in the coolant and affecting the fuel integrity through interactions between spallation products and the blanket.[216] The corrosive effects of LBE and the fact that some of the spallation products are volatile may increase the desirability of having two separate flow loops for the target and the reactor coolant.[217] More information concerning the potential health effects of LBE is provided in Chapter V.

Sub-critical Reactors

All accelerator-based systems use a sub-critical reactor for transmutation. As discussed in Appendix C, the criticality state of a system is described by a factor k, which is known as the multiplication factor. A reactor with a k of 1 is exactly critical (i.e. each fission produces exactly one more fission). This is the level maintained for conventional fission reactors. A k above 1 is super-critical (this is always the case in nuclear weapons and is the case when a reactor power level is increasing). When k is less than 1, the reactor is sub-critical.[218] The sub-critical reactors proposed for ATW range from a k of 0.95 to 0.98 (with many proposals opting for a k around 0.97). This means that the reactor must have an external source of neutrons to maintain the chain reaction.[219]

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[215] NAS-NRC 1996, pp. 246-247, Levanov 1999, and ATW Roadmap 1999e, p. 150 and B-9. As noted by the NAS-NRC, "multiple nuclear reactions involving high energy neutrons in the target produce a host of nuclides, many of which are radioactive." Only some of those have been identified here. Also, the lead-bismuth would be considered "mixed" waste and would be regulated under both hazardous waste regulations (lead and bismuth are toxic heavy metals) and radioactive waste regulations. A calculation for a 1 MW test chamber operating for six months showed that the total specific activity of the lead-bismuth could reach ~500 Ci/kg, reducing after five years to 2.7 Ci/kg (Yefimov et al. 1999). See also Shubin et al. 1999.

[216] ATW Roadmap 1999e, p. 3-3

[217] Levanov et al. 1999, p. 10 and ATW Roadmap 1999e, p. 3-3

[218] Makhijani and Saleska 1999, p. 40-42

[219] The degree of dependence of sub-critical systems on external neutrons can be understood in the following way. For a k of 0.95, every 100 fissions results in another 95 fissions. Thus, the neutrons from spallation, when the reactor is operating at constant power, are responsible for 5 out of every 100 fissions.

There is a fundamental choice to be made in designing the system as to the level of subcriticality of the transmuter core. A lower k means that the core is further from criticality which requires a higher power accelerator and reduces the economics of operating the system (since more power has to be used for the accelerator as opposed to generating revenue). As k reaches 1, the accelerator power required drops significantly.[220] This has a number of implications for design and operation of the system, including the degree to which the safety improvements supposed to be provided by ATW will be realized.[221]

In a critical reactor the multiplication factor is adjusted to increase or decrease the power level, shut down the reactor, or to keep a constant power level, by the use of neutron absorbing materials, including control rods and, in some reactors (PWRs), chemicals added to the moderator or coolant (called a chemical shim).[222] Initially, the reactor is fueled in such a way that there is more fissile material present than necessary to achieve criticality. However, the chemical shim and full insertion of the control rods prevent fission. The reactor is then brought to its operating critical level by controlling the amount of shim and the position of the control rods. As the fuel reaches higher burn-up levels and fission product poisons build-up, the control rods are gradually adjusted to allow the reactor to remain critical.[223] At a certain stage it becomes necessary to re-fuel the reactor.[224]

Similarly, in sub-critical systems the multiplication factor is adjusted for the same purposes. As the actinides fed into the transmutation reactor are fissioned, it then becomes necessary to increase the number of spallation neutrons produced by increasing the current of the accelerator. This adjustment of current can be quite large and has implications for both the cost of the system,[225] as well as for safety (this is discussed further below).

The other alternative, in order to minimize the need to adjust accelerator current, is to include a fertile fuel in the transmutation reactor such as Th-232 or U-238, which would produce fissile U-233 or Pu-239. These fissile isotopes would buildup, as the initial load of actinides is fissioned, helping to maintain the reactivity. This is identical to how current critical reactors operate. However, this would defeat some of the goals of implementing a transmutation system (see below and Chapter V for a further discussion of the Th-U fuel cycle proposed in some ATW schemes).

In other words, the reactor core itself still dominates the fission chain reaction production. See NAS 1995, p. 197 for a simple explanation.

[220] ATW Roadmap 1999e, p. 3-2. It should be noted, that as the reactivity of the core changes during operation of the reactor, the k of the core will change. This must be factored into safety analyses and may necessitate control rods and other reactivity control mechanisms (ATW Roadmap 1999e, p. 6-15)

[221] It should be noted, that as the reactivity of the core changes during operation of the reactor, the k of the core will change. This must be factored into safety analyses and may necessitate control rods and other reactivity control mechanisms (ATW Roadmap 1999e, p. 6-15)

[222] Lamarsh 1983, p. 275

[223] Each fission produces two (and in rare cases three) fission products. Some of these fission products have large cross-sections for absorbing neutrons and thus have an impact on the multiplication factor. Lamarsh 1983, p. 276.

[224] See Section 7.5 of Lamarsh 1983.

[225] Bowman 1998, p. 540.

Because sub-critical reactors do not rely upon the fuel to maintain a constant reactivity and because of their supplemental source of neutrons, they are more flexible in terms of the radionuclides that can be irradiated. It is believed that a wider range of actinides can be placed in a sub-critical reactor for transmutation. Furthermore, the supplemental neutrons can also be used to transmute technetium-99 and iodine-129 (which can create problems in critical reactors, which depend on internally generated neutrons to maintain criticality). However, this must be weighed against the increased complexity of the system and the need to integrate a number of sub-systems and some of the other problems posed by sub-critical reactors as discussed elsewhere.

Sub-Critical Reactor Safety

The sub-criticality of the reactor core and the dependence on accelerator produced neutrons fundamentally affects the safety of these reactors. Additionally, some designs are relying on what are generally termed "passive safety features" (such as cooling by natural convection) which do not require active controls such as adjustment of control rods. Such reactor designs are often described as "inherently safe." The concept of "inherently safe" will be addressed in Chapter V. While it is true that sub-critical reactors may provide some safety advantages over critical reactors, it is equally true that they still have safety problems, including some which are unique to these systems. This section will provide an overview of the safety features of sub-critical reactors. The focus in this section is on the reactivity control of the reactor as this is the major difference between a sub-critical and a critical reactor. It should be noted that sub-critical reactor design and operation will still have to account for other types of accident scenarios that are common to both types of reactors. Specifically, loss of coolant or loss of flow accidents in which heat cannot be conducted away from a hot reactor core (for example, due to blockage of a coolant channel) are still an issue with sub-critical reactors.

There are a number of positive safety improvements that sub-critical reactors can provide over critical reactors. Systems can be designed to automatically shut-off the accelerator (or block the accelerator beam) and therefore stop the production of neutrons. In some ways this acts in a similar fashion to the rapid insertion of control rods in a critical reactor. According to some sources, this may be even faster than control rod insertion.[226]

One of the key issues in determining reactor safety is to examine how a reactor responds to an increase in reactivity (called a "reactivity insertion"). A "reactivity insertion" results in an increase in power and temperature of the reactor. Sub-critical reactors can have an advantage over critical reactors when it comes to "reactivity insertions" because the increase in power and temperature tends to be slower. This provides a longer period of time for safety systems to come into play before the fuel melts, a dangerous potential result of unchecked increases in reactor temperature.[227] This difference in response to reactivity insertions is very important when fertile-free fuels are

[226] Ritter et al. 1999, pp. 358
[227] Ritter et al. 1999, p. 358

used, since some of the mechanisms intrinsic to the fuel that are ordinarily relied upon to counter the effects of reactivity insertions may not be present (see below).[228]

However, the claim of inherent safety needs to be taken with some caution for both generic and specific reasons. It also should be noted that designs for accelerator based systems are still quite preliminary and much of the needed safety analysis has not been conducted. As the MIT review panel has noted, "The evaluations to date indicate that the proposed design has important, if not unique safety features which preclude criticality and overpower transients. Nevertheless, this aspect must be subjected to intense scrutiny during future evaluations. It would be important to ascertain more fully the range of reactivity swings from mechanistic and hypothetical conditions in order to preserve the desired sub-criticality conditions."[229] In fact it is not clear at all that there are safety features which preclude accidental super-criticality. Furthermore, under conditions in which coolant has been lost the core can melt. This results in fuel slumping and reconfiguration of the fissile material. A melted core could become critical without the external neutron source.[230]

One new safety problem introduced by coupling accelerators with sub-critical reactors is the need to control the neutron production by the accelerator/target system so as not to drive the reactor to a super-critical state. Two examples of this problem will illustrate the point that any new reactor design, while solving one problem, may create new ones. The first example arises from the need to maintain a constant reaction level in the reactor. As the fissile transuranic elements are fissioned the reactivity of the core will change. For a variety of reasons, including economical operation of the reactor, it is preferable to maintain the reactivity at a constant level. With ATW systems there are three possible means to do this (the first two are common practice in critical reactor systems while the third is unique to accelerator based systems):

- Use more frequent fuel shuffling and reloading. However, greater fuel handling increases costs and risks to workers.
- Have some form of neutron absorber material. As in critical reactors this would be in the form of a burnable poison or control rods. This then raises the possibility of the sub-critical reactor having some of the same safety failure modes as a critical reactor (e.g. control rod ejection).
- Raise the current of the accelerator. Increasing the current results in more neutrons available to fission the actinides. Thus an accelerator can have a quite large operating range, starting at a much lower current than the maximum and slowly ramping up the current over the course of reactor operations. This could cause an accidental super-criticality if the accelerator is at full power when fresh fuel is in the reactor.[231]

A second example of a potential new safety problem with ATW systems as compared to current critical reactors, is the change in fuel composition due to repeated

[228] ATW Roadmap 1999f, pp. 29-31. Fertile-free fuels, such as zirconium-plutonium fuels do not produce more fissile radionuclides upon irradiation. This is in contrast to fertile fuels such as uranium-plutonium in which more plutonium is produced when U-238 nuclei capture neutrons.
[229] Kazimi et al. 1998, p. 2
[230] ATW Roadmap 1999f, pp. 30-31
[231] Broeders 1999, p. 348-251. Also Bowman 1998, p. 540 and ATW Roadmap 1999f, pp. 29-30.

irradiation and reprocessing of the fuel. The composition of the transuranic fuel will change over the lifetime of the reactor, resulting in a relative buildup of americium and curium. The change in fission characteristics of the fuel, as well as the change in spontaneous neutron production, will need to be accounted for in the design of the reactor, choice of control systems and operations, and in fuel management strategy.[232]

The use of fertile fuels in ATW systems will also be an issue. The production of fissile U-233 from Th-232 will actually increase the reactivity of the core at some points during irradiation (in contrast to the discussion above which assumed non-fertile fuels). Depending on the initial core reactivity, this could result in a core that is no longer sub-critical according to some calculations.[233] Thorium systems will also have to account for the decay of protactinium-233 (the intermediary radionuclide in the production of U-233 from Th-232) after shutdown of the reactor. During reactor operations equilibrium is reached between Pa-233 production and decay and some of the resultant U-233 is being fissioned. However, after reactor shutdown, all of the Pa-233 will be decaying to U-233, thereby increasing the reactivity of the core.[234]

In addition to new safety concerns raised by the combination of accelerator and sub-critical reactor, there are certain features of ATW that reduce some of the safety features of critical light water reactors.

Neutron Kinetics: The change in fuel composition results in a change in the neutron behavior of the fuel. As noted in the ATW Roadmap report:

> The neutron kinetic behavior of source driven systems differs in fundamental ways from that of critical systems. Source transients can be initiated by changes in accelerator beam characteristics or by changes in the geometry, temperature, or composition of the spallation target. System neutronic response to both source transients and reactivity transients is governed by prompt neutrons, and -- depending on the subcriticality level and the magnitude of the perturbation -- both the magnitude and the spatial shape of the fission power may be very sensitive to such changes. Moreover, the fertile free fuel composition in ATW leads to a low delayed neutron fraction and Doppler coefficient. A detailed understanding of the space-time dynamic behavior of ATW during operational transients and potential accident sequences is needed to confirm operability and safety and to formulate requirements on system monitoring, control and safety protection systems.[235]

Reactivity Feedbacks: Critical reactors rely on reactivity feedbacks to reduce power in abnormal situations. For example, a temperature rise in the coolant/moderator of an LWR due to an increase in reactivity decreases the density of the coolant. This results in lower neutron moderation, which decreases the reactivity of the reactor. This negative feedback is crucial to reactor safety. In other cases, the reactivity may be positive (e.g. an increase in temperature results in an increase in reactivity which of course results in an

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[232] ATW Roadmap 1999f, p. 30

[233] Buccafurni and Orazi 1999, p. 411. Other calculations presented at the Mol Conference for Th-Pu and Th-U systems show an increase in reactivity of the core at some point during irradiation (which would be accompanied by a decrease in accelerator current). However, others do not show the core multiplication factor exceeding 1 (for the proper accelerator input). For example, see Fernandez et al 1999.

[234] NAS 1995, p. 203. This effect of this post-shutdown reactivity change will be greater for larger neutron fluxes, as are expected in ATW systems.

[235] ATW Roadmap 1999f, p. 30

increase in temperature). Such positive feedbacks must be counteracted by other negative feedbacks.[236]

These feedback mechanisms depend heavily on neutron spectrum, coolant, moderator, and fuel type. The ATW systems proposed in the U.S. Roadmap for example are less sensitive than current LWRs to reactivity feedbacks in cases where there is a loss of coolant. This increases the requirements for having prompt and effective shutdown mechanisms for the neutron source.[237]

The fertile-free fuel also reduces the effect of one beneficial feedback mechanism called Doppler broadening. Doppler broadening arises because the thermal motion of the atoms in the fuel increases probability of interaction between the neutrons and the nuclei in the fuel (both the fissile nuclei and the non-fissile nuclei). However, in light water reactors where U-238 forms the bulk of the fuel, the dominant effect is to increase the absorption of neutrons by the U-238. A rise in temperature due to an increase in reactivity results in increased thermal motion and a corresponding larger neutron capture cross section for U-238. Therefore fewer neutrons are available to fission the fuel resulting in a decrease in reactivity and therefore a decrease in temperature. Thus, this feedback serves to limit the effects of a reactivity increase in conventional LWR fuel, helping to bring the reactor back to normal operation. Doppler broadening feedback occurs fairly quickly. However, in fuels without any fertile elements (i.e. fuel without isotopes which absorb neutrons relatively readily without fissioning), the Doppler feedback mechanism can result in an increase in the fission rate. This is because the plutonium and other actinides are the only nuclei present to interact with the neutrons and the result is an increase in the fission rate and a related increase in the temperature. Thus, this important negative feedback becomes a positive feedback. This must be compensated for by other feedback mechanisms. As a result an upper limit is set to the amount of fissile fuel in such reactors in order to keep the overall fuel temperature feedback mechanism (both Doppler broadening and other feedbacks) negative.[238]

This is not to say that these reactivity issues cannot be mitigated through the design of the core and the operation of the reactor. All of the reactivity feedback mechanisms will have to be taken into account in determining how the core is fueled and the level of the accelerator current at different times during reactor operation. However, this does illustrate the point that every technology based upon large quantities of radioactive materials, including fissionable material, has an inherent safety issue associated with it and that no reactor system is completely immune from major accidents.

In considering the safety of sub-critical reactor systems it is important to note that the risk of a criticality accident is only one of many potential problems which can occur in a nuclear reactor. According to Dr. Lawrence Lidsky of the Nuclear Engineering Department of the Massachusetts Institute of Technology (MIT):

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[236] For more information about reactivity feedbacks, see Bodansky 1996, pp. 191-193 and Lamarsh 1983, pp. 306-316.
[237] ATW Roadmap 1999f, p. 30-31
[238] Bodansky 1996, pp. 191-192

> A major "selling feature" of the ATW schemes is the sub-critical nature of the system. However, this is a misleading distinction. When considering the total risk of nuclear accidents, criticality accidents are low on the list as compared to meltdown accidents. Sub-critical systems can actually be more dangerous than conventional reactors if, as is often the case, there are more subsystems that can fail or initiate failures, and fewer backups. . Probabilistic risk analysis is a complex art, requiring a deep understanding of possible accident initiators and accident progression, and the ATW design is far too rudimentary at this time to apply this powerful tool. However, it is clear that the currently envisaged ATW systems are more complex than fission reactors, have more accident initiators, and many fewer backup safety systems. On the basis of the comparison, I would *guess* that the probability of a significant accident in a Rubbiatron would be orders of magnitude greater than that of a modern fission reactor. The importance of sub-criticality is frequently overestimated by physicists ignorant of the engineering aspects of nuclear reactor design. Regulators know better.[239]

As noted above, one of the accident scenarios which remains of issue, whether the core is sub-critical or not, is proper cooling of the core.

In addition to the reactor physics problems that may complicate sub-critical reactor safety there are other safety issues unique to accelerator based programs. The problem of beam trips and reliability was discussed above. This can result in thermal stresses that can affect the structural integrity of reactor components in addition to causing power peaking in the fuel.[240] Another potential problem would be the use of lead-bismuth eutectic for coolant and spallation target. One of the reasons the United States set aside its efforts to develop liquid lead based coolants for its nuclear program is the corrosive effects of the lead (as well as problems related to oxygen balance and the production of radioactive polonium through neutron capture).[241] International transmutation efforts that look to make use of liquid lead or lead-bismuth eutectic are using Russian experience with this technology to guide them.[242]

Another problem is the durability of the beam window. In order to separate and protect the accelerator tube (which is a vacuum through which the proton is accelerated before striking the spallation target), a window is placed between them. This beam window would experience severe conditions related to the proton flux through the window, the neutron irradiation of the window, thermal stresses, and the corrosive effects

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[239] Lidsky 2000

[240] See above and see Takizuka 1999 , p. 384

[241] ATW Roadmap 1999a, p. A-13

[242] The Russian solution to this problem is a combination of material choices and careful control of the coolant chemistry. The oxygen levels need to be precisely controlled in order to have sufficient oxygen to form a protective oxide layer on the coolant structure. The materials for the coolant system are chosen to enhance the protection offered by the oxide layer. However, too much oxygen would lead to the formation of lead oxide. The Russian experience, while the most extensive, is still modest. A total of 80 reactor-years of experience is reported for LBE in comparison to 260 reactor-years for sodium and 100 reactor-years for helium, neither of which has been widely implemented. (see ATW Roadmap 1999f, pp. 10-15) Furthermore, it should be noted that the Russian experience arises from its classified submarine propulsion program and as such would have to be extended to cover spallation targets in addition to reactor coolant.

of the LBE.[243] This is further exacerbated by the desire to have as thin a window as possible in order to minimize proton energy loss.[244]

As was noted in a paper by Los Alamos National Laboratory scientists, "Subcriticality does not make ATW by definition 'safer' than critical reactors. Rather, subcriticality facilitates tasks that would be exceedingly difficult or inefficient in critical systems."[245] The degree to which these tasks are facilitated and the new safety problems that arise in ATW systems are still being determined. All of the possible accident scenarios must be considered before determining the safety of these systems and it is premature to make claims of enhanced nuclear safety at this time.

Specific Accelerator Based Research Programs

Significant research and development work must occur for any transmutation scheme to be put into place. This is particularly true of accelerator based programs which would need to break new ground in a number of fields. Accelerators would have to be developed that could meet the high current and reliability requirements necessary for transmutation. The spallation target system would have to be developed and tested to ensure that it could meet all of the requirements. Lead-bismuth research would have to be advanced beyond its use as a coolant for submarine reactors. The sub-critical cores would have to be designed and assessed for safety. Further development of the reprocessing operations would have to occur in order to meet the high separation requirements necessary for transmutation. Within each of these areas are a number of challenges and problems which may or may not be able to be overcome. Furthermore, integration of all of these subsystems will pose a challenge in and of itself. Materials will have to be assessed to ensure that they can withstand the extreme conditions that would be experienced, particularly in the spallation and reactor regions. Beyond the engineering and materials science issues are the fundamental physics of some of the processes that will have to be better understood and modeled (e.g. reactor models that account for a supplemental source of neutrons).[246]

It would appear that only a sustained and costly research and development program would be able to answer some of the questions. Those research and development programs are currently underway or being put into place in a number of countries.

[243] ATW Roadmap 1999f, pp. 46-47, ATW Roadmap 1999e, p. 3-3, 3-4. Development and testing of candidate materials for the window would be part of the research and development program for ATW.
[244] ATW Roadmap 1999a, p. A-13
[245] Venneri et al. 1998, p. 4
[246] For details on the outstanding issues related to the different technologies for ATW (accelerators, reactors, reprocessing), see the supporting documents for the ATW Roadmap.

The four most important research and development efforts in this area are

- the U.S. ATW Roadmap effort because it has identified many of the outstanding technical hurdles and is the most comprehensive description of how ATW would be used for the existing waste of a country;
- the Rubbia Energy Amplifier because it represents the most advanced proposal for cyclotron based sub-critical transmutation;
- the French program because of the French Parliament's mandate to explore transmutation and because of France's current use of MOX; and
- the Japanese program because of ongoing active Japanese research in this, and other advanced nuclear power, proposals to solve Japan's energy security problems.

U.S. ATW Roadmap

The FY 1999 Energy and Water Appropriations Act included the requirement that the U.S. Department of Energy conduct a road-mapping study on the development of accelerator transmutation of waste. The resulting report provides some of the most detailed information available on the current state of knowledge of the various components needed to implement transmutation. It is also provides detail on how a specific transmutation program might be implemented by providing a scenario to transmute the entire stock U.S. civilian spent fuel.[247] The report did not, however, contain a systematic assessment of whether transmutation is the most reasonable alternative and is incomplete in its assessment of the consequences of transmutation. This is discussed further in Chapter V.

The ATW program proposes to separate and transmute the actinides, technetium, and iodine. Strontium and cesium would be separated, but not transmuted. Instead they would be sent to the repository. Uranium would also be separated for re-use or disposal as low-level waste. The ATW would also produce electricity of which 85-90% would be sold.[248]

The Roadmap identifies a both baseline and alternative technological options.[249] The following are the baseline options identified in the Roadmap. The ATW system uses a 45 MW linear proton accelerator based on the one under development for tritium production. The separations technologies chosen are a combination of aqueous (UREX) and pyrochemical processing for the LWR spent fuel and a pyroprocessing system for the ATW fuel. The spallation target and coolant are LBE and the fuel is a solid metal fuel made from the actinides (15%) and the zirconium (85%) from the original cladding of the spent LWR fuel.[250] The technetium and iodine are fabricated into transmutation assemblies to be put into the sub-critical reactor. The neutron spectrum is a fast one, though an end-of-life burn-down scenario has been proposed in which the spectrum

[247] In addition to the Roadmap report, there were six supporting documents produced which focus specifically on accelerators, separations, cost, target/blanket, the geologic repository performance and systems scenarios and integration.
[248] ATW Roadmap 1999, p. A-2
[249] See the ATW Roadmap 1999e report for details about the technology options for all of the technology categories (e.g. separations, accelerator, coolant, etc.)
[250] Venneri et al. 1998, p. 3

would be thermalized at the end of the life time of the reactor in order to take advantage of neutron capture resonances.[251]

While much of the system is based upon technologies in use there are significant components which require development. As noted above, the accelerator requirements are above the current state of the art. Pyrochemical processing has worked on the laboratory scale but has never been used commercially and will require considerable development work.[252] Metallic fuel is not in widespread use and in the United States has been confined to experimental reactors. Significant development and testing work will be required. LBE as a spallation target in addition to coolant will have to undergo further development. Finally, integration of all of these subsystems will be a very difficult task. The process by which safety of a novel system will be established is likely to be complex and drawn out.

In addition, there are a number of regulatory, institutional and public policy issues that would have to be addressed. These would include overturning the current policy to forego commercial reprocessing, public concerns over safety, financial responsibility for both development and eventual deployment, the need to ensure proper regulatory oversight, export control questions, and many others.[253]

According to the DOE, a full-scale program would take approximately 118 years (including development) and 8.5 ATW stations in order to transmute 87,000 metric tons (MT) of spent fuel.[254] The demonstration phase would end and full-scale operations would begin in 2028 (assuming an immediate start to the program in this fiscal year). Each station would consist of two linear accelerators and eight sub-critical reactor cores producing 840 MWt.[255] During those 90 years for actual transmutation the amount of TRU would be reduced from 905.5 tons to at least 2.4 tons.[256] The TRU inventory plus 3,000 tons of fission products and 13,000 tons of zirconium would be sent to the repository. Over 82,000 tons of uranium would also have to be disposed of. As a result, there would be an increase, or at least no net change, in the mass of materials to be

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[251] Venneri et al. 1998, p. 4

[252] ATW Roadmap 1999d, p. 13.

[253] See ATW Roadmap 1999e, Chapter 7.

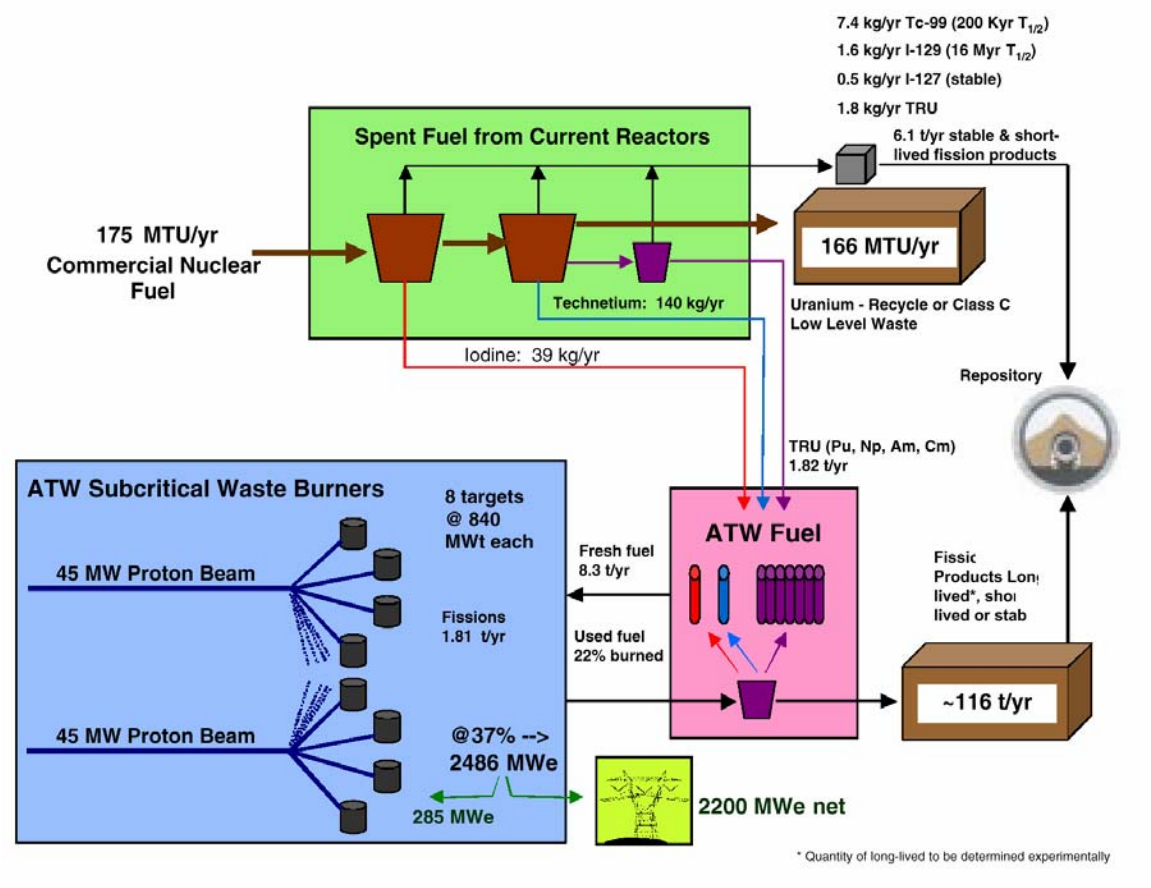
[254] ATW Roadmap 1999a, p. A-1. This amount of spent fuel is the projection for LWR reactor spent fuel production under current operating licenses and assumes no new reactors are constructed and operated.

[255] ATW Roadmap 199a, p. 7-1, 7-2. Each unit (two accelerators, eight reactors, plus separations facilities) would operate for approximately 60 years (76 years if one includes design and decommissioning). Initially, there would be a demonstration facility that would be upgraded to a half-size production facility. R&D and demonstration would be completed by Year 28. New eight unit power stations would come on-line in a staggered manner with all power stations operating concurrently approximately in Year 50. The first station would be shut down in year 100 and the last station shut down in year 118. It should be noted that there seems to be some discrepancies on the exact number of plants required to transmute the 87,000 ton inventory. ATW Roadmap 1999a p. 7-2 and p. A-1 indicate that there would be eight plants (with the initial demonstration plant upgraded to a full facility). However, ATW Roadmap 1999e p. 5-2 (as well as ATW Roadmap 1999d, p. 38 and others) indicate that 8.5 stations would be required. For the purposes of analysis, this report assumes 8.5 stations are required.

[256] ATW Roadmap 1999d, p. 38. This includes only the amount of transuranics in high level from reprocessing, according to the flowsheet on p. 38. However, it is necessary to note that there would a significant amount of transuranics left in the last reactor core once it is shut down. It is unknown at this time what will become of that material.

disposed of. The system as it is currently proposed is illustrated in Figure 19. Figure 20 provides a more detailed preliminary flow diagram of the materials balances for the ATW system and shows the amount of materials processed and the waste produced.

Figure 19: U.S. ATW Roadmap Proposed System

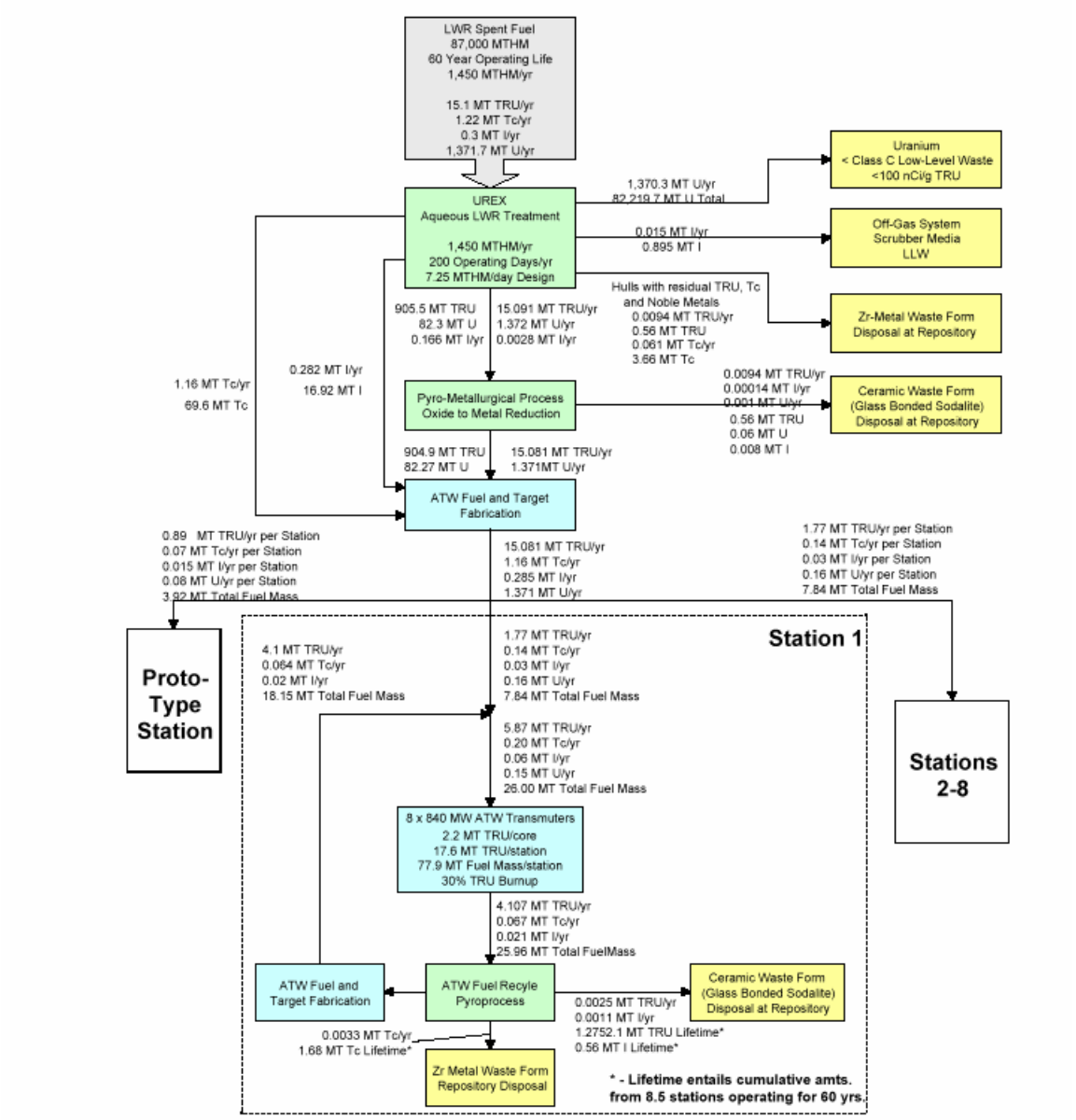


Source: ATW Roadmap 1999a, p. E-10

During operation each station will have an inventory of 17.6 tons of TRU.[257] Each station will also process 4.1 tons per year of TRU in the ATW pyroprocessing facility. The burn-up rate of the actinides is not clear. According to the Roadmap report it is 22%, but according to the supporting technical document it is 30%.[258]

[257] ATW Roadmap 1999d, p. 38
[258] See ATW Roadmap 1999a, p. A-1 for the 22% figure and ATW Roadmap 1999d, p. 37 for 30% figure.

Figure 20: U.S. ATW Roadmap Preliminary Materials Flowchart



Source: ATW Roadmap 1999d, Figure 10, p. 38

The roadmap also includes a cost estimate for the R&D program, development of the technology and full-scale implementation of transmutation for the projected 87,000 MT of spent fuel. Table 11 provides a summary of those costs (this does not include the costs for the repository, which are discussed below). The roadmap proposes to start with a $281 million, six-year, research and development program. This program would not only continue technology development work but also would undertake studies and of existing technologies Systems studies, which would evaluate differing deployment options would be undertaken and the particular mix of technologies to be used would be determined. While the initial focus has been on accelerator based systems, according to

the DOE the systems and trade studies would not rule out any particular technology.[259] Thus, the final analysis may favor implementing critical reactor technologies and aqueous processing. It may also favor other variations on the ATW program just described (e.g. using sodium coolant instead of LBE or using a cyclotron instead of a linac).

Table 11: Summary of Estimated Undiscounted ATW System Life-Cycle Costs (billions of 1999 dollars)

System Element	R&D	Demo	Implementation			Total
			Capital	Operating	D&D	
Accelerators	0.17	2.5	11.2	44.4	0.6	58.8
Transmuters	1.03	2.1	30.2	49.4	3.1	85.8
Separations	0.50	2.2	9.0	40.5	1.0	53.3
ATW Fuel Fabrication	--	0.6	2.1	40.7	0.2	43.6
Site Support	--	1.0	1.0	30.6	0.1	32.7
Retrieval/Transportation/Disposal	--	0.1	--	4.2	--	4.3
Integration	0.07	0.9	--	--	--	0.9
Subtotals	1.77	9.4	53.5	209.8	5.0	279.4

Reproduced from Table 4.1 of the ATW Roadmap 1999g.

As is discussed below in Chapter V, the electricity revenues are not expected to recoup all of the costs of ATW deployment (once a discount rate is applied to account for differences in the value of money over time, the shortfall would be about $14 billion). In addition, there is the question of the cost of a repository. Since ATW will not eliminate the need for a repository as was hoped for by its early proponents, the costs of a repository will still be incurred. In other words, the ATW costs are in addition to the repository costs, not instead of the repository costs. Furthermore, ATW costs include an extra $4.3 billion for retrieval, transportation, and disposal on top of the base costs of the repository.

In Fiscal Year 2000, a further $9 million was provided to conduct trade studies and perform some experiments related to ATW. The ATW program is planning to develop a program plan on how to proceed, should a decision be made to proceed with research and design activities. That program plan will be sent to Congressional committees and the Office and Management and Budget. According to the FY2001 DOE Budget request, no further money is requested for ATW. However, it should be noted that the initial $4 million for the Roadmap and this year's $9 million were not included in any budget request from the administration and were added by Congress.[260] Thus, it is possible that further funds for ATW could be added in this year's appropriations bill. Furthermore, once the program plan is sent to the various parties, it is possible that either the Administration or members of Congress will seek further funds.

While the focus of US efforts has been ATW, there have been attempts in the last two years in Congress to establish an Office of Spent Nuclear Fuel Research. It was proposed in the past two nuclear waste bills vetoed by the President and an identical provision is in the Energy Security Act of 2000, introduced by Senators Lott and

[259] Personal notes of Hisham Zerriffi at the December 1999 meeting of the Department of Energy, Office of Nuclear Energy, Nuclear Energy Research Advisory Council (NERAC).
[260] FY 2000 and FY 2001 Department of Energy Congressional Budget Requests for Nuclear Waste Disposal.

Murkowski. This Office would require reprocessing and transmutation research on a full suite of technologies. This would presumably include aqueous reprocessing systems and critical reactors.[261]

Rubbia's Cyclotron System

Carl Rubbia, a Nobel laureate of the CERN in Geneva, has been promoting a concept called the "Energy Amplifier." Cyclotrons would be used to accelerate protons to the 1.0 GeV energy level. A system using three, progressively larger, cyclotrons has been proposed. The third cyclotron could also be replaced by a linear accelerator. The proton beam would have a current of 12.5 mA. The module would produce 1500 MWt of thermal energy, and a three module plant would altogether produce 2000 MWe of electrical energy from the 4500 MW of thermal energy.[262]

Initially, the Energy Amplifier was intended to be a replacement for light water reactors that would utilize a thorium-uranium fuel cycle operating as a breeder reactor. An initial load of fuel containing thorium and enriched uranium would result in the production of fissile U-233 from the Th-232. The U-233 would be extracted by reprocessing and used as fuel in subsequent cycles once sufficient quantities were produced. Subsequently, the Energy Amplifier has been proposed as an accelerator transmutation of waste system that would be fueled initially with actinides from light water reactors. The U-233, which would build up in the fuel, would be separated and stored until the task of transmuting the actinides was complete. The stored uranium would then fuel the reactors.

The most detailed examination of the Energy Amplifier for transmutation purposes appears to be in the context of the Spanish nuclear power industry.[263] A total of five EAs would be required to transmute the actinides from the nine light water reactors in use in Spain. The program would take 37 years (not including R&D, demonstration, etc.). The accelerator would be a three stage system starting with a linear injector, a six sector cyclotron and then either a LINAC or a 10-12 sector cyclotron. The energy of the protons would be 1.5 GeV and the current would be varied between about 7 and 16 mA for control of reactivity (the design calls for 4 mA above the maximum required current to provide a margin for operation of the accelerator so the maximum current of accelerator would be 20 mA).[264] The EAs would be metal-fueled fast neutron systems using molten lead as a coolant. The reprocessing system would be pyroprocessing. In addition to the TRUs, the plan also proposes to transmute Tc-99 and I-129.[265] The demonstration phase for this proposal would extend to 2010-2015 with the production phase ending around 2045-2050.[266]

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[261] See for example, the Nuclear Waste Policy Amendments Act of 2000 (S. 1287, vetoed by the President) and the National Energy Security Act of 2000 (S. 2557).
[262] Rubbia et al 1997a, p. 187
[263] Rubbia et al. 1997b
[264] Rubbia et al 1997b, pp. 53-54
[265] Rubbia et al 1997b, p. 1
[266] Rubbia et al 1997b, p. 5. A 10 MW demonstration facility is also being proposed by Spain (ATW Roadmap 1999a, p. 3-2)

Perhaps most significantly, the proposal claims that the need for a geologic repository would be completely eliminated. The uranium would be separated for reuse. The transuranics, technetium, and iodine would be put in the EA.[267] Long-lived radionuclides would be diluted in order to meet the requirements for disposal as low-level waste, which set certain concentration limits. This would include all of the important long-lived fission and activation products (with the exceptions of technetium and iodine, which would be put in the transmutation reactor and those emitted during reprocessing).[268] Strontium and cesium, two of the more significant medium-lived radionuclides, would be put in storage for hundreds of years and also possibly diluted. Storage and dilution would be conducted with the goal of meeting the low-level waste disposal requirements. The transfer from above-ground storage to waste disposal would occur anywhere from 150 to 640 years after the start of transmutation operations depending on the level of dilution and whether the waste is disposed of as Class C or Class A waste. Rubbia et al. also note that with sufficient dilution the fission products could be disposed of immediately as Class C waste.[269]

As will be discussed in Chapter V, these claims need to be approached with a great deal of skepticism. Repeated reviews of transmutation have reaffirmed the need for a repository. Claims that the EA would completely burn the transuranics through repeated recycling[270] ignore practicalities like process inefficiencies and hold-up in facilities which will always leave at least a residual amount of transuranics to be disposed of. Furthermore, as with most other transmutation proposals, the focus is on the radiotoxicity of the transuranics rather than the dose estimates from the repository which indicate that other long-lived fission products play a large role (see Chapter V). As for proposals to store fission products for long periods of time and/or to dispose of fission products and residual waste as "low-level" waste, this raises serious concerns. These are addressed in Chapter V.

French program

In December of 1991, the French Parliament passed a law which, in part, required R&D on partitioning and transmutation. While initial work focused exclusively on critical reactor based systems, French researchers have recently begun to include ATW research. A number of organizations within France are involved, both on their own projects and in collaboration with each other and international research programs. Studies are being conducted at the systems level as well as on specific topics such as sub-critical reactor physics, accelerators, spallation, and separation chemistry.[271]

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[267] Rubbia et al 1997b, p. 2

[268] Rubbia et al. 1997b, p. 2, 29. The low-level waste disposal regulations of the United States (10 CFR 61) are used in this paper. It is not clear why the U.S. regulations are used when the proposal concerns disposal of waste in Spain. U.S. low-level radioactive waste regulations include three classes A, B, C, with progressively more stringent requirements. Waste is classified according to the concentration of specific radionuclides. This is discussed further in Chapter V.

[269] Rubbia et al. 1997b, pp. 29-32

[270] Rubbia et al 1997b, p. 1

[271] Salvatores, Schapira, and Mouney 1997 , pp. 423-429

The French program remains focused heavily on the use of critical reactors (as discussed in Chapter III). Implementation of accelerator based sub-critical systems would likely occur in a multi-level system. Such a system would include conventional LWRs, LWRs fueled with MOX, fast reactors, and ATW. The French experience with reprocessing and the desire to continue the use of critical reactors indicates that aqueous separations would continue even if pyroprocessing were to be implemented for ATW fuel.

Current projects include collaboration with Japan, Russia and the United States for a test loop using LBE. The LBE loop is being constructed and will be cold tested in Russia and then shipped to Los Alamos National Laboratory for hot testing (with radioactive materials) with the resulting data shared among the partners.[272]

Japan

As part of its OMEGA program, which encompasses a number of potential transmutation technologies, the Japanese government is sponsoring research on all aspects of separations and transmutation. Research is being conducted on new fuels based on nitrides for actinide irradiation, aqueous and pyroprocessing, accelerators, sub-critical reactor cores and integrated systems. The Japan Atomic Energy Research Institute (JAERI) has done design studies on an ATW system. The reference system uses a 1.5 GeV, 33 mA linac, a tungsten spallation target and a sodium-cooled sub-critical core (820 MWt). An LBE cooled system is also being developed as an alternative option. The fuel is a nitride-based fuel and the reprocessing facilities would use pyroprocessing. The system would produce 270 MWe of electricity of which 100 MWe would be used to run the accelerator.[273] It is not clear why this accelerator would only be used to drive a single core, while a similar accelerator in the U.S. ATW plans would drive four sub-critical cores. However, it should be noted that without running multiple cores, the economic operation of such a facility would be even more difficult.

Other Efforts

In addition to the programs discussed above and other national programs in South Korea, the Czech Republic, Sweden and other countries, there is internationally coordinated work being done on transmutation. As mentioned above, the programs in Japan, France, and the U.S. are funding a test loop for LBE, which is being constructed in Russia, and there are other bi-lateral and multi-lateral collaborations. International organizations and entities are also sponsoring work in this area. The Nuclear Energy Agency of the Organization for Economic Cooperation and Development (OECD/NEA) has held regular meetings and recently released a comprehensive status report on transmutation.[274] Conferences have been held in the United States, Russia and Europe and the IAEA has sponsored meetings and produced reports. The European Union is finalizing what is called the Fifth Framework Agreement, which will provide over $15

[272] Herczeg 1999
[273] Takizuka et al 1999a, Mukaiyama et al 1999, Suzuki et al 1999 and Kubota et al 1999.
[274] OECD-NEA 1999a and 1999b

million in funding in addition to national funds being expended. This will support research in a variety of areas.[275]

[275] ATW Roadmap 1999a, p. 3-3,3-4

Chapter V: Implications

Chapters I-IV provided an overview of the waste management problem, transmutation as a solution, and the specifics concerning the technologies that would be used for transmutation. This chapter will address the implications of transmutation in order to determine whether it would meet its stated objectives and what liabilities it may have. Transmutation programs would obviously have an impact on nuclear waste management, but that impact would not be necessarily be a positive one. Transmutation involves a continuing need for a repository, the possible implementation of long-term aboveground storage for some materials, and potential increases in other types of radioactive waste due to processing. It also would involve significant costs, both monetary and otherwise. This chapter will review issues related to nuclear reactor safety, cost, environmental safety and health, proliferation, and the development of new nuclear

fuel cycles which may arise as a result of transmutation being implemented widely. The chapter will close with an assessment of the impact of transmutation on nuclear waste management.

The Results of Transmutation

In order to better understand the implications of transmutation it is useful to review exactly what transmutation can accomplish for the various different types of radionuclides present in high level nuclear waste. First, however, a brief summary of U.S. nuclear waste regulations is provided in order to place these results in context.

Radioactive Waste Classification

In the United States there are a number of different categories of radioactive waste. There are a number of problems with the existing classification system, including the fact that it is not based on the longevity of the waste and hence is not always protective of future generations. The issues involved are complex and beyond the scope of this report, however, they are discussed in other works by the Institute for Energy and Environmental Research and elaborated on a little in the section later in this chapter on low-level waste.[276]

The United States has three nuclear waste classifications of relevance to transmutation:

High Level Waste: High Level Waste (HLW) can refer to either spent fuel from nuclear reactors or the liquid waste from reprocessing operations, which is subsequently solidified. The latter is sometimes referred to as high level liquid waste (HLLW). Both forms of HLW must be disposed of in a geologic repository according to US regulations. The only repository site currently being considered in the United States for HLW disposal is Yucca Mountain in Nevada.

Transuranic Waste: Transuranic waste is a category that consists of waste contaminated with heavier radionuclides. The term "transuranic" literally means "above uranium" and refers to all of the elements above uranium on the periodic table (i.e. with atomic numbers greater than 92). However, U.S. government regulations define "transuranic waste," also called "TRU waste," as any waste containing transuranic elements with half-lives greater than 20 years and in concentrations above 100 nanocuries per gram. The transuranics include plutonium, americium, and curium (though not all isotopes meet the 20 year half-life threshold). Most transuranic waste in the US is the result of processes involving the production of nuclear weapons and can include both homogenous contaminated waste (e.g. soil) and bulk items such as boots and gloves. Transuranic waste must be disposed of in a geologic repository. Currently, in the United States, transuranic waste is slated to be disposed of in the Waste Isolation Pilot Plant in New Mexico.

[276] Readers interested in further information concerning waste classification issues are referred to Makhijani and Saleska 1992 and Ortmeyer 1997.

Low Level Waste: This category of waste is a catch-all category and is essentially defined as that waste which is not high-level, transuranic, or uranium mill tailings. Within the low-level waste category there are three sub-classes (Classes A, B, C) with Class C waste being submitted to the most stringent requirements of the three. All three classes can be disposed of in shallow land burial though the disposal requirements (e.g. level of containment) vary by Class. Waste is put in a class according to the concentration of specific radionuclides specified in a chart contained in the Code of Federal Regulations. Long-lived radionuclides include carbon-14, technetium-99, iodine-129 and the transuranics. Short-lived radionuclides include cobalt-60, strontium-90, and cesium-137. Some radionuclides of importance to transmutation systems, such as uranium and cesium-135, are not included in the classification system and their status is in question.

In other countries, a similar three level waste classification system is established with high, intermediate, and low-level waste categories. However, the difference is that the waste classification is according to the longevity and hazard of the waste, rather than according to the process that produced the waste or arbitrarily limited to certain radionuclides (as is the case with transuranic waste in the United States).[277]

Medium-Lived Fission Products

All transmutation schemes separate out the medium-lived fission products, particularly strontium and cesium. As discussed in Chapter I it is infeasible to transmute Sr-90 and Cs-137, which dominate the radioactivity of the medium-lived fission products, because of both small neutron absorption cross-sections and the presence of other isotopes. There are, thus, two possible fates for the medium-lived fission products. Many transmutation proposals would store the medium-lived fission products above ground for approximately 500 years.[278] This is to reduce the radioactivity level of Cs-137 and Sr-90 such that they could be disposed of as Class C waste in shallow land burial. As discussed below, low-level waste burial technology has a history of failure in protecting the environment and would endanger water resources. The other possibility would be to place these wastes in the repository along with the residual actinides and long-lived fission products or in their own repository. Sending this waste to a long-term repository would be in accordance with current US regulations which treat all waste from reprocessing of irradiated spent fuel as high level waste to be sent to a repository.

[277] This issue is dealt with in detail in Makhijani and Saleska 1992.

[278] Ordinarily, the standard rule of thumb is that a radionuclide has decayed away to negligible levels after ten half-lives. This corresponds to a factor of about 1000 reduction in the amount of the radionuclide. For Sr-90 and Cs-137, a factor of 1000 reduction takes around 290 and 300 years respectively (ten half-lives). However, this reduction may not be sufficient given the extremely large amounts of Sr-90 and Cs-137 in the waste. Therefore, a more stringent requirement, corresponding to a reduction of 10^5-10^6 may be more appropriate. For the same radionuclides it would take approximately 400-600 years (13-21 half-lives) to get this level of reduction. This longer time matches some scenarios being proposed (for example, Rubbia et al. 1997b).

Long-Lived Fission Products (LLFP)

The only long-lived fission products widely proposed for transmutation are Tc-99 and I-129. Tc-99 appears to be the least difficult of the fission products to transmute. It can be extracted during reprocessing and fabricated into a metal target. It is transmuted into a stable ruthenium isotope, which is also a metal with a high melting and boiling point.

On the other hand, the transmutation of I-129 is somewhat uncertain. In some cases, particularly if aqueous reprocessing methods are to be used, the I-129 would be released to the environment during reprocessing. This is the current practice, for example in France. However, even with the capture of the highly volatile iodine, its fabrication into targets and irradiation is not as simple an operation as for Tc-99. Not only is there the problem of chemical instability of the target, but the product of I-129 transmutation is a noble gas (Xenon-130) which will have to be vented to avoid pressure build-up in the target.[279] Avoiding such pressure build-ups, which can be a safety and operational concern, complicates the design of the target. Another problem with I-129 transmutation is the possibility of I-129 gas being vented from the reactor if the reactor has an increase in temperature.[280]

While some have suggested transmutation of Cs-135, this would be impractical for reasons already discussed. The rest of the long-lived fission products would be sent to the repository without transmutation. In certain repository scenarios many of the fission products considered to pose the highest dose risk cannot be transmuted (see below).

Actinides

One of the main reasons cited for transmutation is to reduce the inventory of actinides due to their high radiotoxicity and proliferation risk. The level of actinide reduction will depend in great part upon the mix of transmutation technologies chosen.

In order to have more complete fissioning of actinides, some form of accelerator based system will be required (either as part of a hybrid scheme or alone). Even with ATW there remains significant amounts of actinides in the waste (at least 2.4 metric tons out of an initial 900 tons will be contained in the high-level waste for repository disposal in the case of the US Accelerator Transmutation of Waste plan).

One method to measure the reduction in the inventory of transuranic materials due to transmutation is to use the "transuranic ratio" (sometimes called the Pigford-Choi equation). The transuranics ratio is defined as[281]:

$$\psi(t) = \frac{\begin{array}{c}\text{Total inventory of transuranics sent to waste disposal in time for the reference} \\ \text{once - through LWR fuel cycle, if no fuel reprocessing, no recycle, and no transmutation}\end{array}}{\begin{array}{c}\text{Total inventory of transuranics at time t in the transmuter,} \\ \text{in its fuel cycle, and in process wastes}\end{array}}$$

[279] OECD-NEA 1999b, p. 53
[280] OECD-NEA 1999b, p. 53
[281] NAS-NRC 1996, p. 59

The denominator (the bottom half of the equation) is relatively straightforward. It is the amount of waste at any given time if transmutation is being undertaken. The numerator is slightly more complicated. It is the amount of transuranics in the waste that would be sent to the repository if transmutation does not occur. However, it is not only the transuranics in the waste to be transmuted. This is because the transmutation reactors also produce electricity for sale. If one assumes that LWRs (which would also produce more transuranics) would otherwise produce the electricity then there is a certain amount of transuranic production that is avoided. The numerator includes both the amount of transuranics to be transmuted and the amount of transuranic production avoided.

Another number that can be calculated is the "depletion ratio" over a given period of time, which is symbolized as $\chi(t)$. This ratio is simply the amount of transuranics sent to the transmutation reactor divided by the amount of transuranics contained in the transmutation system (transmuter, reprocessing and associated systems, and the waste). This ratio is lower than the transuranic ratio since it does not include the amount of transuranics that would be produced if LWRs were generating the same amount of electricity as the transmutation reactors.[282]

The National Research Council, in its 1996 report on transmutation, presented a number of calculations of the transuranic ratio and the rate of transmutation for various transmutation scenarios. The transuranic ratio is of course sensitive to both the types of transmutation reactors used as well as whether transmutation is implemented in the context of constant production of nuclear energy or a phaseout of nuclear energy. The NAS-NRC panel presented calculations for both constant power and declining power scenarios and for the use of Advanced Liquid Metal Reactors, Light Water Reactors, and Accelerator Transmutation of Waste.[283] The results indicate that to achieve any significant reduction of transuranics would require a significant long-term commitment. As noted by the panel:

> If overall TRU ratios of the order of 100 are desired to benefit waste disposal, the ratio must apply to the entire national system of nuclear power generation. Any of the transmutation scenarios considered here would require commitments to construct and operate the transmuter system and its later-generation replacements for long periods of time, of the order of centuries for declining power and many centuries to millennia for constant transmuter power.[284]

It should also be noted that the actual amount of transuranics left after transmutation is higher than might be indicated by the transmutation ratio because it includes the amount of transuranics that would be produced if LWRs produced the equivalent amount of energy. This can be illustrated using the results of the most recent ATW roadmapping effort in the United States. According to the Roadmap the initial inventory of transuranics from LWRs is reduced from 905.5 metric tons to somewhere in the range of 2.5 metric tons plus the residual amount left in the last transmuter (which could be in the range of 1.5-6 metric tons).[285] This would imply therefore a depletion ratio of 105-225

[282] NAS-NRC 1996, p. 59
[283] NAS-NRC 1996, pp. 58-71
[284] NAS-NRC 1996, p. 71
[285] This is a highly simplified calculation based on the following assumptions. According to the Roadmap, as each ATW station is shut down, its inventory of TRU will be distributed to the remaining stations. As

over the course of 118 years. If the amount of equivalent energy that the ATWs produce in that time period were instead generated by LWRs, then there would be an additional 275 metric tons of transuranics.[286] Thus the transuranic ratio would be 140-295. Of course, this calculation is not very definitive as it includes a number of assumptions, both those made in the Roadmap and those made by the authors. For example, the Roadmap assumes that each reprocessing step will achieve a process loss of 10^{-4}, which is a significant improvement over current commercial reprocessing. If such a low process loss cannot be achieved, then the depletion ratio and the transuranic ratio could be significantly lower. In any case, it should be noted that the amount of transuranics to be sent to the repository must still be measured in terms of tons of material rather than grams or kilograms and still represents a significant amount of material.

Other Radionuclides

Carbon-14: Carbon-14 is produced primarily by neutron reactions on nitrogen and is thus a neutron activation product. It has a half-life of 5,730 years and poses a particular problem because of its potential for incorporation in biological molecules, replacing the stable carbon with one that will decay into a new element. Under certain repository conditions it can become dissolved in groundwater and be released in gaseous form. In particular, C-14 is a problem for unsaturated repositories where it is more easily released. An EPA Advisory Panel found that release of half of the C-14 from an unsaturated U.S. repository would lead to a small increase to the individual dose (below the regulatory limit) but a large overall population dose over 10,000 years such that it would result in an excess of 4,000 cancer deaths.[287]

While most transmutation proposals do not specify how they will handle the C-14, two solutions have generally been put forth. The first is to trap the carbon-14 in the off-gases of the reprocessing facilities, convert it into a physically stable form and then dispose of the final waste form in a repository. The second possibility is to continue the general current practice of releasing the C-14 to the air during reprocessing. As transmutation would involve a significant increase in reprocessing operations, this would greatly increase the amount of C-14 released. According to the National Research Council, the release of C-14 from reprocessing of U.S. spent fuel would be within current

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the final ATW station reaches the end of its lifetime, there arises the question of the final inventory in the reactor cores. If the reactor cores were progressively shut down until only one core remains and that core is then run for a typical cycle (i.e. there is no deep burndown) the remaining TRU inventory would be 1.5 metric tons. The core which would initially be fueled with 2.2 metric tons of transuranics and would achieve a 30% burnup (ATW Roadmap 1999d, pp. 37-38). However, each accelerator powers four reactor cores. If for some reason, all four cores must remain operational, then all four cores would have a residual inventory of 1.5 metric tons for a total of six metric tons. However, this all speculative (for example, it does not account for the possibility of a deep burndown scenario in which the final core achieves higher than 30% burnup) and this calculation should be considered a rough approximation.

[286] Each ATW station produces 2110 MWe net. We have assumed, as the NAS did, 1,395 MWe pressurized water reactors operating at 33 MWd/kg (Megawatt days per kilogram of heavy metal, a measure of the amount of energy generated from a unit of fuel) and 0.80 capacity factor producing 359 kg/yr. would be used to provide the equivalent amount of energy (NAS-NRC 1996, p. 64). Thus each ATW station produces the same energy as 1.5 LWR. For 8.5 stations operating for 60 years each the total equivalent TRU production from the LWRs would be 275 metric tons.

[287] EPA 1993, p. 21.

regulatory limits and would only be a fractional addition to the global inventory of C-14 due to natural processes.[288] However, this seems to ignore the findings of the EPA Advisory Panel since release of this amount of C-14 into the air would be expected to result in a significant number of cancer deaths over the period of time during which C-14 decays. The number of excess cancer deaths could be expected to be double or more that found by the EPA panel.[289] The EPA panel noted the difficulties in resolving the issue of individual risk versus population risk for radionuclides such as C-14 which become uniformly dispersed once they are released (unlike some other radionuclides, once C-14 is released it mixes in the atmosphere relatively rapidly).[290]

Uranium: The vast majority of the mass of spent fuel is composed of uranium. Currently, both depleted uranium (from enrichment of natural uranium) and extracted uranium are treated as a low-level waste or as a product to be used.[291] The question of disposal of the vast quantity of uranium that would be separated from spent fuel as a result of transmutation programs is a very important one. The U.S. ATW Roadmap calls for it to be treated as a Class C waste or stored (presumably for re-use).[292] However, if the goal is protection of the environment and public health then both the DOE and the Nuclear Regulatory Commission must consider another option. The regulations governing both agencies call for geologic disposal of all wastes containing transuranic radionuclides at concentrations above 100 nCi/g.[293] IEER has recommended elsewhere that depleted uranium be treated in the same manner as TRU waste (currently TRU waste is being sent to the recently opened Waste Isolation Pilot Plant) due to the fact that an analysis of the physical properties of depleted uranium showed it should meet the criterion for TRU waste.[294] Irradiated uranium is even more radioactive than depleted

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[288] NAS-NRC 1996, p. 100

[289] The EPA panel result was based on a release of half of the C-14 contained in 70,000 metric tons of spent fuel (the limit for the first U.S. repository) over the course of 10,000 years and used a figure of 1 Ci of C-14 per metric ton (EPA 1993, p. 19). Reprocessing of the full 87,000 metric tons of spent fuel expected in the US from current reactor operations would result in all of the C-14 from that spent fuel being released immediately (if no attempt is made to capture the C-14 during reprocessing). Furthermore, the NAS-NRC panel uses a figure of 1.5 Ci of C-14 per metric ton (NAS-NRC 1996, p. 26).

[290] EPA 1993, p. 29

[291] In the United States for example, uranium is not listed in the tables of specific radionuclides for determining the class of low-level waste (10 CFR 61.55). It is therefore treated, by default, as a Class A low-level waste. Depleted uranium is also used by the military (e.g. for armor-piercing rounds) and for other commercial purposes.

[292] ATW Roadmap 1999a, p. 1-2

[293] 40 CFR 191 for the DOE and 10 CFR Part 61.55 Table 1 for the NRC (this sets the Class C limit to <100 nCi/g). Both regulations cover only alpha-emitting radioisotopes with atomic number 93 or higher (e.g. higher than uranium on the periodic table). However, the regulations do differ slightly. Under DOE regulations, the radionuclides have to have a half-life greater than 20 years while under NRC regulations the cutoff is greater than 5 years. Furthermore, while DOE regulations specify TRU disposal in a geologic repository, the NRC regulations for greater than Class C waste call for geologic disposal unless an alternate plan is approved by the NRC.

[294] Makhijani and Makhijani 1996. The major isotopes of uranium are all alpha-emitters with half-lives well in excess of 20 years. While it is true that plutonium is more radioactive (due to its shorter half-life) in a direct comparison of plutonium and uranium (e.g. assuming the same amount of each element), this has been factored into this analysis since it is a comparison of the radioactivity per unit mass of the waste. See Makhijani and Makhijani 1996 for a more detailed explanation of the comparison of TRU waste and uranium waste.

uranium and should also be treated as TRU waste. While the lower limit on TRU waste is 100 nCi/g, our calculations show that the re-extracted uranium would have a concentration of nearly 1400 nCi/g.[295] Table 12 provides a summary comparison of the specific activity of the uranium extracted from LWR spent fuel in comparison to depleted uranium, transuranic waste and uranium ore. As can be seen, the extracted uranium has a much higher specific activity than either the depleted uranium or the limit for transuranic waste.[296]

Clearly, uranium extracted as part of reprocessing operations should not be disposed of in shallow land burial as a low-level waste. Rather, it should be treated in the same manner as transuranic waste, which is currently required to be disposed of in a geologic repository. The fact that uranium is not included in either the low-level waste or transuranic waste regulations is a loophole in the law that should be closed. There is no scientific basis for treating this waste as a Class C or lower waste. Given the level of radioactivity of the extracted uranium, there is also no scientific basis for reusing it. While this analysis has focused on U.S. waste classifications, the situation in other countries is believed to be similar. All countries which currently separate uranium as part of reprocessing for MOX, or who would implement reprocessing for transmutation, should also treat their re-extracted uranium as a waste comparable to plutonium waste.

Historically, some of this re-extracted uranium was sent to enrichment facilities for re-enrichment as light water reactor fuel. Recent investigations in the United States have shown that workers and the public at these facilities, such as the one in Paducah, Kentucky, were put at risk due to the presence of both plutonium and fission products, such as Tc-99, in the re-extracted uranium. These elements are not present in the fresh uranium usually processed by these facilities.[297] Re-use of the uranium in reactor fuel would also be counterproductive if the goal is to reduce the level of actinides since new actinides would be produced, possibly even resulting in a net gain in actinides (see Chapter III). This uranium would also pose an increased risk if used for the other military or commercial purposes for which depleted uranium has been used.

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[295] The specific activity of extracted uranium was found by weighing each isotope's specific activity by its percentage in typical spent PWR fuel as presented in NAS-NRC 1996, Table 2.2, p. 25. This does not take into account the radioactivity due to any TRU contamination or Tc-99 contamination as is commonly found in reprocessed uranium. The specific activity of the final waste form may be less (since it is unlikely to be pure uranium metal), and would be still far higher than the 100 nCi/g limit. For example, we calculate that as an oxide (U_3O_8) the waste would be approximately 1200 nCi/g.

[296] Table 1 modified from Makhijani and Makhijani 1996 by adding calculated values for extracted uranium.

[297] For example, see the series of articles by Joby Warrick of the *Washington Post* and U.S. DOE Office of Oversight, Environment, Safety, and Health, *Phase I Independent Investigation of the Paducah Gaseous Diffusion Plant (Environment Safety and Health Practices 1952-1990)*. October 1999 and Phase II report of February 2000.

Table 12: Specific Activities of Various Chemical Forms of Extracted LWR Uranium, Depleted Uranium, TRU Waste and 0.2% Uranium Ore

Chemical form	Specific activity, nCi/g
Extracted LWR uranium metal (U)	1400
Extracted LWR uranium oxide (U3O8)	1200
Depleted uranium metal (U)	360
Depleted uranium oxide (U3O8)	300
Depleted uranium tetrafluoride (UF4)	270
Depleted uranium hexafluoride (UF6)	240
Transuranic activity in TRU waste	100 (See note 2)
0.2 % uranium ore	4 (See note 3)

Notes for Table 12:

1. Specific activities of the four forms of uranium [U, U3O8, UF4, UF6] have been rounded to two significant figures, and that of uranium ore to one significant figure.
2. The minimum limit of 100 nanocuries/gram of transuranic elements for waste to be classified as TRU waste includes only those isotopes of transuranic elements with half-lives greater than 20 years. The most common isotope in TRU waste that is eliminated from the counting in this way is plutonium-241, which has a half-life of 14.4 years. However the decay product of plutonium-241, americium-241 is included in TRU waste because it has a half-life of about 432 years. All the uranium isotopes dealt with in this comparison have half-lives far longer than 20 years.
3. The specific activity of 0.2 percent uranium ore shown includes all decay products of uranium-238 up to and including radium-226, assuming they are in secular equilibrium with uranium-238. Radon-222 and its decay products are not included.

Neutron Activation and Spallation Products: There are currently no plans to transmute either neutron activation products from current and new reactors or spallation products from accelerator based systems. Some neutron activation products (e.g. carbon-14, chlorine-36) are considered to be significant contributors to air and groundwater pathway doses for certain repositories. These radionuclides are long-lived and may move into the environment rapidly. Their small cross-sections, difficulty in extraction or capture during reprocessing, and/or the presence of stable isotopes prevent their transmutation. No transmutation scheme has addressed this set of radionuclides. More neutron activation products may be produced in accelerator based systems due to the high neutron levels in the target and reactor regions (as well as activation of the accelerator structure).

Accelerator based systems will add the problem of spallation products in the neutron source. These are the residual radionuclides after the neutrons have been spalled. The radioactivity from these spallation products poses a problem for both occupational doses and for waste management.

Liquid and Solidified High Level Waste

There is an open question as to whether existing quantities of waste from prior reprocessing operations would be transmuted. This waste has arisen from both military and commercial reprocessing using PUREX and its predecessor processes. There are large volumes of liquid high level waste as a result, some of which has been solidified through vitrification (essentially encasing it in glass logs). This waste contains a significant amount of fission products as well as actinides, in particular neptunium, americium, and curium. Table XX of Chapter I, comparing spent nuclear fuel and

commercial liquid high level waste in the United States and France, illustrates the magnitude of the problem.

For example, at the Hanford Site in Washington State, one of the sites of plutonium separation for the United States nuclear weapons program, there is an estimated 206,000 cubic meters of reprocessing waste contained in large storage tanks. The radioactivity of this waste is about 200 million curies with most of that radioactivity coming from Sr-90 and Cs-137, two radionuclides that are not candidates for transmutation.[298] In all, of the 70,000 metric tons of high level waste slated to be sent to the Yucca Mountain repository, approximately 7,000 is from defense high level waste and defense spent fuel.[299]

While processes are being developed (for example in France and Japan) to extract these actinides and some fission products after an initial PUREX extraction of plutonium, it is not clear whether these would be applied only to waste generated in the future or would also be applied to existing waste. If existing liquid waste were not processed and transmuted this could significantly constrain the supposed benefits of transmutation in a number of ways. For example, an upper limit would be placed on repository expansion due to transmutation of actinides and storage of medium-lived fission products. This is due to the fact that the heat of the waste limits the capacity of repositories and the reprocessing waste contains high heat medium-lived fission products and higher actinides that may be difficult and expensive to remove. It would also mean that potential dose reductions due to the transmutation of commercial spent fuel would not be as significant due to the contribution of reprocessing waste to the dose (this is discussed further below in the section on repository doses).

One of the major impediments to extracting Tc, I, or the minor actinides from previous reprocessing waste will be cost. The only method to recoup some of these costs is through the sale of electricity. However, the plutonium and overall fissile content, of the reprocessing waste may be far too low to justify the costs of extraction. This was the conclusion reached by the National Academy panel as well as by proponents of transmutation.[300] The National Academy Panel noted that in comparison with the amount of TRU in spent commercial fuel (and its relative purity) "there appears to be no justification for performing expensive further separation processes so that a fraction of the defense waste TRUs can be transmuted rather than sent to the repository."[301]

Reactor Safety

Safety is a key issue in determining the costs of any new nuclear system, whether it is a new conventional light water reactor or an accelerator based sub-critical reactor. A number of designs for reactors have been proposed which are described as inherently safe, often due to their reliance on what are described as "passive safety systems."

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[298] Fioravanti and Makhijani 1997, p. 148 and NAS-NRC 1996, p. 89. For more information about the Hanford waste tanks and problems in dealing with the waste contained within them, see Fioravanti and Makhijani 1997, Chapter 3 and NAS-NRC 1996, Chapter Five.
[299] DOE 1999b, p. Figure A-2, p. A-8
[300] NAS-NRC 1996, p. 97
[301] NAS-NRC 1996, p. 97

However, both the critical and the sub-critical reactors proposed for transmutation pose certain safety risks which result either from the use of fuels containing plutonium and minor actinides or from new design features or both.

It should also be recognized that changing certain design features in order to make a reactor "safer" such as moving to a sub-critical reactor, does not eliminate other serious safety considerations that remain. For example, Loss of Coolant Accidents (LOCA) are a major safety challenge for all nuclear reactors. Loss of coolant can led to fuel meltdowns with serious consequences, no matter what the level of criticality, since even when a reactor is not operating at full power there is considerable heat from the radioactive decay of the fission products. The coolant must still remove this heat. A break or blockage of a coolant pipe can result in a failure to adequately remove that heat. This is what led to the partial meltdown of an experimental breeder reactor in Idaho in 1955. In fact fuel melting or other reconfiguration of the fuel can change the reactivity of the fuel above the criticality level, even in a reactor that is designed to be sub-critical.[302]

Critical Reactor Safety

A change in the fuel of critical reactors must be accounted for in the safety analysis of critical reactors, as discussed in Chapter III. In particular, the use of plutonium and other actinides as fuel changes the behavior of the fuel due to differences in the fission and neutron absorption of uranium, plutonium, and the minor actinides. Changes in the rate of fission, the number of delayed neutrons, the reaction to temperature changes in the fuel, and the reactivity during off-normal conditions (e.g. loss of coolant) act to decrease the safety margin of critical reactors fueled with plutonium or minor actinides as compared to uranium fuels.[303]

In order to mitigate the effects of plutonium and minor actinides on reactor safety, there is a need to (a) increase the use of neutron absorbers (e.g. control rods), (b) to limit the amount of transuranics loaded into the reactor (which therefore sets limits on the transmutation rate) and (c) set limitations on the form and placement of transuranics in the reactor (i.e. whether they should be loaded in the core or in the periphery of the reactor and whether transuranics should be incorporated into the fuel or fabricated into targets).[304]

Sub-Critical Reactor Safety

As discussed in Chapter IV, sub-critical reactors change certain key features about reactor operations with an effect on the safety of the reactors. In some cases this effect can be beneficial. Because the core of the transmuter would be sub-critical, there would be a difference in the response time of the reactor under certain conditions. This longer response time could help in control of the reactor. The reliance of the reactor on the

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[302] Bodansky 1996, pp. 212-213. Lamarsh 1983, pp. 605-607. ATW Roadmap 1999f, p. 31

[303] Of course, it should be recognized that even in reactors initially fueled with UO_2 a significant portion of the energy of the reactor is from fission of plutonium. However, there are significant differences in the amount of plutonium and other actinides present in the fuel from neutron absorption by uranium as opposed to a reactor deliberately fueled with plutonium.

[304] See Chapter III

spallation neutrons also provides another method for reactor shutdown in an emergency and it has been proposed that accelerator shutdown may be quicker than control rod injection in a critical reactor.

However, as was noted in Chapter IV, sub-critical reactors also provide new challenges for reactor safety, which must be addressed. These include beam window durability, power peaking problems due to accelerator beam trips, over-power transients due to accelerator current control, and the potential for fuel melting. These are just a few examples of the complex set of problems that must be considered when designing a nuclear reactor, even if it is sub-critical.

"Inherently Safe" Reactors[305]

Accelerator based sub-critical reactors are only the latest in series of reactor designs which have been promoted as being "inherently safe." As can be seen by the above discussion of safety issues with both critical and sub-critical reactors, it is impossible to completely eliminate all reactor safety concerns through design of the reactor. In fact it is extremely problematic to apply the term "inherently safe" to nuclear reactors. Consider the following three facts:

1. All reactors, whether critical or sub-critical, contain large quantities of radioactive and toxic materials.
2. All reactors rely on a process (fission) that has the potential to be self-sustaining and out of control (super-criticality) and which generates a large amount of heat and energy which can result in melting of components (including fuel) and explosive releases of energy.
3. All reactor designs to date have experienced at least one major accident.[306]

In essence nuclear reactors are inherently unsafe as they involve dangerous materials in a process with the potential for serious accidents. This is not to say that certain reactor designs are not safer than others. It is very obvious that the design of current light water reactors in use in North America or Western Europe are safer than those of the RBMK reactors built in the former Soviet Union. However, it is an issue of relative risk and the mitigation of the risk. All safety systems designed for nuclear reactors, both passive and active, are included to counter the inherently unsafe nature of nuclear reactors. If these reactors were truly inherently safe they would not require safety systems at all.

ATW systems are a perfect example. They have certain features that provide them with safety advantages over critical reactor systems (e.g. the ability to shut off neutron production and the fact that the sub-critical core takes longer to be affected by a reactivity insertion, thus providing safety systems with a longer time period in which to respond). However, ATW systems also require both active and passive safety systems to be included in the design and in some cases these safety systems are new. Failure of these safety systems could result in a catastrophic accident. ATW systems also result in new safety problems for which mitigation measures must be devised (e.g. systems to

[305] For a more complete discussion see Makhijani and Saleska 1999, Chapter 7

[306] See Makhijani and Saleska 1999, Table 7, pp. 152-153 for information on some of the reactor accidents to date.

avoid high currents with high reactivity fuels). It is unclear how the safety of such a complex system at full scale would be tested.

It must also be recognized that safety issues are not limited to the design of the reactor. There is a significant human factor involved both on the level of operator error and on the organizational level. Thus, a number of accidents in the nuclear arena which should not have occurred according to the design of a facility or according to the established procedures did occur because safety systems were circumvented or because the management was lax concerning safety or other human error. The criticality accident at Tokaimura in October 1999 is just one example whereby existing safety procedures were not followed in order to maximize production. In prior nuclear reactor accidents both design and operation problems have been at fault (and in some cases both). For example, the Chernobyl reactor accident was due to both problems with the design of the reactor and in the manner in which the reactor was operated.[307]

In conclusion, it is necessary to recognize that nuclear reactors are extremely complex pieces of machinery, which are then operated by fallible human beings. Furthermore, the consequences of any reactor accident, no matter the initial cause, could be severe due to the potential for release of significant quantities of radiation.

Cost

Given that implementation of transmutation will require a significant expansion of the nuclear infrastructure it is necessary to examine the financial implications of such programs. This is particularly important given historic cost escalations of nuclear power in general, and reprocessing operations and breeder reactors specifically. It should be noted that transmutation, due to the higher heat and radioactivity levels, would also result in increases in fuel fabrication and handling costs that will have to be factored into the costs of these fuel cycles. These high costs, coupled with the low throughput, will result in significantly higher unit costs for fuel fabrication. For example, the OECD estimated fuel fabrication costs for the LWR fuel cycle to be in the range of $232-$464 per kg of uranium (adjusted to 1999 dollars).[308] MOX fuel is estimated to be about four times more expensive to fabricate and just adding neptunium homogeneously to the MOX fuel will further increase costs by an estimated 20%.[309] By comparison, the ATW roadmap estimates fuel fabrication costs to be at least $2,700 per kg processed.[310]

Unfortunately, complete life cycle cost estimates are not easy to find, particularly for critical reactor based transmutation proposals. In addition, cost estimations and comparisons are made somewhat more difficult because different transmutation proposals assume differing scenarios for the future of nuclear power. For example, some transmutation proposals for multi-reactor transmutation systems are based on the assumption of continuing use of these reactors in equilibrium rather than their use to

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[307] Bodansky 1996, pp. 222-223

[308] OECD-NEA 1994, p. 37

[309] OECD-NEA 1999b, p. 37. This OECD-NEA report uses a figure of $275-300 per kg of uranium for ordinary light water reactor fuel fabrication costs. This is roughly comparable to the earlier OECD-NEA report.

[310] ATW Roadmap 1999g, p. 4.19. This figure is for the eighth ATW station. The first ATW station will have fuel fabrication costs estimated at over $11,000/kg because it will not operate at full capacity during the demonstration phase and operates for a longer period of time.

process only existing LWR spent fuel. The ATW Roadmap provides the most complete cost estimate available for the full life-cycle costs of a transmutation program to handle a specified amount of LWR fuel and is summarized herein. First, however, a review of reprocessing, breeder reactor, and pressurized water reactor MOX fuel cycle costs is provided.

Reprocessing Costs

The National Research Council report on transmutation has provided an excellent review and analysis of the cost of fuel reprocessing.[311] The unit prices of reprocessing (the prices charged to reprocessing customers per kilogram of material processed) based on actual industrial experience range from $600 to $1,400/kgHM (1992 dollars).[312] The most recent OECD/NEA report on fuel cycle costs set a cost for reprocessing at $770/kgHM.[313] As noted, by the panel, these are for aqueous reprocessing facilities that are designed solely to separate uranium and plutonium after one pass through the reactor. For the removal of all transuranics for transmutation, the costs of aqueous reprocessing would be even higher.[314] Part of that increase comes from the need for more advanced reprocessing techniques and some comes simply from higher shielding requirements due to environmental, safety, and health considerations. For example, the OECD-NEA review of transmutation included the results of a European Union study that estimated the increased fuel cycle costs due to advanced reprocessing and transmutation. For the most advanced reprocessing facilities necessary to separate americium, neptunium, curium technetium, and iodine the construction costs would rise by fifty percent over a conventional reprocessing facility.[315] Just separating americium and neptunium could raise reprocessing costs by approximately 15% and overall fuel cycle costs by anywhere from 10% to 50%.[316]

Based on the prices quoted above, and a cost estimate by the OECD/NEA, the NRC Separations Technology and Transmutation Systems (STATS) panel also made an estimate of reprocessing costs in the United States, which does not currently have commercial reprocessing capabilities. These results are shown (1992 dollars) in Table 13. These costs do not include the costs of decommissioning the facility.

[311] NAS-NRC 1996, pp. 112-117 and Appendix J
[312] NAS-NRC 1996, p. 431. This is based upon prices reported for THORP in the U.K. and UP3 in France.
[313] OECD-NEA 1994, p. 12. The OECD-NEA figure is 720 ECU per kilogram. In order to be consistent with the National Research Council panel estimates we have used their conversion rate of 1.07 ECU per dollar (NAS-NRC 1996, p. 433).
[314] NAS-NRC, p. 117
[315] OECD-NEA 1999b, pp. 311-312
[316] OECD-NEA 1999b, pp. 311-312

Table 13: Calculated Unit Costs for Conventional Aqueous Reprocessing, 900 Mg/yr., U.S. Financing

Plant Owner/Operator	Unit Cost of Reprocessing, $/kg
Government	800
Utility	1,300
Private Industry	2,100

Source: NAS-NRC 1996, Table 6-9, p. 116. Note: One megagram (Mg) is equivalent to one metric ton of heavy metal (MTHM).

The panel compared these unit costs with those of the DOE laboratories and contractors that have been studying transmutation. The costs of aqueous reprocessing in most studies range from $237 to $600/KgHM.[317] Costs of pyrochemically processing LWR fuel were estimated at around $350/kgHM.[318] The more recent estimates from the ATW Roadmap are discussed below.

The cost estimates provided by the DOE and contractors to the STATS panel, as well as other industry studies, were significantly lower than both historical operating experience would indicate or the estimate by the STATS panel. The conclusion of the panel was that "The latter [DOE and contractor estimates] are so much lower than those estimated in the present study that there is good reason to question the validity of all the recent U.S. estimates for the cost of reprocessing LWR spent fuel."[319]

Underestimates of reprocessing costs for transmutation are not limited to the United States. In applying the cyclotron based Energy Amplifier concept to the case of Spain, Rubbia et al. also seem to underestimate reprocessing costs.[320] Unfortunately, they do not provide a unit cost. However, they estimate capital costs for the pyroprocessing facility to cost $1 billion and Operations and Maintenance costs to be $0.1 billion/year.[321] Given their assumptions that it will take 37 years to transmute the 9,628 tons of Spanish spent fuel, this corresponds to a unit cost of $488/kgHM.

Breeder Reactor Costs

Many transmutation proposals rely on the use of reactors based upon those designed to breed plutonium. These breeder reactors were of varying design but were all supposed to be able to eventually produce as much or more plutonium than they consumed and therefore produce their own fuel. This was envisioned to be the future of nuclear power, particularly when it was thought that uranium resources were scarce. With only a few exceptions these reactor designs were based on the use of fast neutrons. It is thus illustrative to briefly examine the cost of breeder reactors as this may give an indication of potential costs for transmutation systems (whether they use critical reactor based on breeders or sub-critical reactors, in which the core is based on breeder reactors).

[317] NAS-NRC 1996, p. 433
[318] NAS-NRC 1996, p. 440
[319] NAS-NRC 1996, p. 117
[320] Rubbia et al. 1997b, pp. 10-14.
[321] Rubbia et al. 1997b, p. 11-13.

After expenditure of tens of billions of dollars in capital costs, most breeder reactor programs have faced serious difficulties and some have been canceled. Overall breeder reactor capital costs have totaled approximately $25 billion (for reactors above 100 MWt).[322] Superphenix, a French reactor, epitomizes the poor performance of these reactors. This 1200 MWe fast breeder reactor was the largest constructed and had approximately the same rated capacity as France's light water reactors. In operation from 1986 through 1997, the reactor actually operated for only 278 days of full power equivalent. This is almost ten times less than a similarly rated commercial nuclear power plant. In addition, the reactor itself was expensive, $9.1 billion (1996 dollars), and the decommissioning and post-operations costs are estimated at another $1.4 billion.[323]

MOX Fuel Cycle Costs

The MOX fuel cycle represents the simplest of transmutation proposals and contains some of the elements found in all transmutation proposals (e.g. reprocessing facilities). There are, of course, major differences between MOX fuel cycles and the more detailed transmutation proposals. For example, plutonium is only passed back through the reactors once before disposal of MOX spent fuel. Furthermore, these fuel cycles currently rely on thermal reactors (such as LWRs) rather than the mix of thermal and fast reactors that characterize most transmutation proposals, or even accelerator based systems in more advanced proposals. Nonetheless, it is useful to compare the costs of once-through and MOX fuel cycles as it can indicate a general trend which would be expected to continue for transmutation proposals (this is further indicated by examining the ATW lifecycle cost estimate, which is presented below).

The Nuclear Energy Agency of the Organization for Economic Cooperation and Development (OECD/NEA) conducted a study that was titled *The Economics of the Nuclear Fuel Cycle*.[324] The main comparison in the study was of Pressurized Water Reactors (PWRs) run in the once-through mode (i.e. spent fuel is directly disposed of without recycling) and in the MOX mode (i.e. reprocessing occurs and plutonium is passed back through the reactors once). The results of that study indicate that the once-through option is 14% less expensive than the MOX option.[325]

Another interesting comparison was conducted by RAND which compared the costs of managing waste from the once-through cycle and the MOX fuel cycle (as well as comparison to what they term the "self-generating recycle" (SGR) which is a fuel cycle in which MOX spent fuel is reprocessed and plutonium is passed through the reactors repeatedly).[326] The purpose of the study was to evaluate the often cited view "that reprocessing helps waste management."[327] As a proxy for manageability, Chow and Jones chose to compare waste management costs, examining the costs of managing waste

[322] See Makhijani 2000. Cost have been converted from local currencies to U.S. dollars (using rates in effect during construction) and adjusted using appropriate price deflators to 1996 dollars.
[323] See Fioravanti 1999 pp. 46-47 and references therein.
[324] OECD-NEA-1994.
[325] OECD-NEA 1994, p. 15
[326] Chow and Jones 1999
[327] Chow and Jones, 1999, p. 4

at each step in the fuel cycle from uranium mining through to spent fuel and high level waste disposal.

The cost estimates for waste management were based on two cost drivers. For high level waste and spent fuel, the cost of waste management is a function of the heat generation of the waste as this establishes the loading of the repository. The findings of the report indicate that the heat of the spent fuel and high level waste is anywhere from 7% lower to 44% higher for fuel cycles based on reprocessing (in comparison to the once-through case). For waste classified as low-level or intermediate-level (which in the United States would be TRU waste), as well as uranium mill tailings, the cost is driven by waste volume. The plutonium fuel cycles generate 5%-9% more low-level waste, 88%-149% more intermediate level waste and 23%-32% less tailings (all comparisons in terms of waste volume generated). As a result, the overall waste management costs are 20%-25% higher for the plutonium fuel cycles in comparison to the once-through fuel cycles.[328]

Repository Costs

The results presented above for MOX fuel cycle costs are significant when considering the potential costs of transmutation proposals. As they all rely on reprocessing operations, with the resultant increase in low-level and intermediate level waste production, the costs for disposal of these wastes can be expected to be higher than for the once-through fuel cycle.

In terms of repository costs, the fact that the cost driver is the heat of the waste, rather than the volume or mass, indicates that transmutation proposals may have a hard time reducing repository costs. First, it should be noted that the timeframe when heat generation is important for setting repository capacity is much shorter than the overall timeframe for assessing repository performance (i.e. decades or hundreds of years rather than thousands or tens of thousands or even millions of years). Second, a significant portion of the heat generation comes from Sr-90 and Cs-137, which are not proposed for transmutation. Third, transmutation also results in the production of higher actinides. The production of isotopes such as Am-241 and Cm-244 counteracts the reduction in heat from the fissioning of plutonium. While this situation is particularly an issue with thermal reactors, there will still be production of higher actinides with fast reactors. Even with accelerator transmutation of waste, the residual actinides can be expected to have a higher proportion of the americium and curium. Thus, the fissioning of long-lived plutonium isotopes may reduce the long-term heat load, but the concurrent production of fission products and shorter-lived actinides increases the short-term heat load. The end result is that cost savings will depend greatly upon the particulars of any transmutation scheme and the heat levels of the resulting waste. It will also depend on how the heat capacity of the repository is determined (e.g. instant measurements of heat at the time of emplacement versus the integration of the heat over a longer time period, say 1,000 years).[329]

[328] Chow and Jones 1999, pp. 1-2
[329] See Chow and Jones 1999, Chapter V.

ATW Roadmap Estimate

The recent roadmap for the U.S. accelerator transmutation program provides the most detailed cost estimate yet available for transmutation. A summary of the costs is provided in Table 14.

Table 14: ATW Roadmap Cost Estimate Summary (billions of 1999 dollars)

System Cost Elements	Undiscounted [330]	Discounted @ 3% for 118 yrs.
Research and Development	1.8	1.4
Demonstration	9.4	5.7
Post-Demonstration Capital Expenditures	53.5	16.1
Post-Demonstration Operations	209.8	29.7
Decontamination and Decommissioning	5.0	0.2
Total System Life Cycle Cost	279.4	53.1
Electricity Revenue	294.9	38.3

Source: ATW Roadmap 1999g, Table 2.1: Estimated Undiscounted and Discounted System Life-Cycle Costs, by Cost Element (billions of 1999 dollars). p. 2-3

Accelerator Transmutation of Waste is expected to result in a net cost of 14.8 billion dollars. This would be *in addition* to the cost of the repository (transmutation would not eliminate the need for a repository). To give some idea of the scale of this endeavor it is useful to compare the expenditures necessary for ATW in comparison to a geologic repository. The total life-cycle cost of disposing of all 87,000 metric tons of spent fuel (plus defense waste) in a repository is estimated to range from $52 -- $57 billion (1999 dollars)[331], about 15 times less than ATW. Of course, electricity revenues would offset the ATW costs, but this comparison does provide a sense of the scale of the endeavor being proposed.

There are a number of factors that could drive ATW costs to be even higher:

- **Electricity Price Assumptions**: The revenue estimate is based upon a price of 43 mills/kWh, which in turn is based on an estimate by the Electric Power Research Institute of fossil-fuel electricity prices.[332] However, as the ATW report notes, the

[330] When considering expenditures and revenues over the course of many years, it is necessary to account for two main factors. The first is the rate of inflation, which changes the purchasing power of money over time. The second is called the discount rate. The discount rate accounts for the fact that if money is invested rather than spent it will earn a rate of return and thus increase the amount of money available later on for the same expenditure. In other words, the value of the money decreases over the years. By spending money, one foregoes the potential income from that investment (Bodansky 1996, p. 318).

[331] TRW 1999, p. 4. It should be noted that this estimate assumes (for the purposes of conducting the estimate) that all 87,000 metric tons would be sent to the Yucca Mountain repository rather than only 70,000 metric tons being sent to the first repository and the rest being sent to a second repository (TRW 1999, p. 2). In addition to the commercial spent fuel, it assumes 2,570 MTHM of defense spent fuel and 20,000 canisters of vitrified high level waste would be sent to the same repository.

[332] ATW Roadmap 1999g, p. 2.2

probability of producing electricity at less than 43.7 mills/kWh is only ten percent and the probability of producing electricity at less than 55.6 mills/kWh is ninety percent. Therefore, "ATW electricity would be unlikely to be competitive with most of the electricity generated commercially in the United States" if current investor-owned utility costs are indicative of future costs.[333] In other words, there is a 10% probability that ATW will generate electricity at a rate expected to be competitive and at that rate revenues will not be sufficient to cover the costs. Currently, fuel cycles using reprocessing are more costly than those based solely on uranium fuel and uranium fuel cycles are more costly than those based on natural gas. Even wind energy is cheaper than MOX fuel cycles.[334] It is highly unlikely that transmutation fuel cycles will ever be cost competitive with either other nuclear fuel cycles or with alternatives to nuclear power.

- **Historical Price Escalations**: Worldwide, breeder reactor costs have escalated (see above). A report by the US General Accounting Office found that many of the U.S. Department of Energy's large projects suffered from budgetary overruns.[335] This problem is continuing as is evident by just two major DOE projects that have recently faced technical difficulties resulting in major cost increases. These are the Defense Waste Processing Facility which processes high-level radioactive waste at the Savannah River Site and the National Ignition Facility, a laser fusion facility at the Lawrence Livermore National Laboratory.[336] NIF's construction cost estimate has now approximately doubled to over $2 billion. Over one billion more dollars will need to be spent on R&D on the project, meaning that the total cost will have approximately tripled.[337] As noted in Chapter IV, the DARHT facility is another example of a DOE project that has far exceeded its original estimated cost. This is a problem that is not new however. A 1981 RAND study concluded that pioneering projects on advanced technologies repeatedly underestimate costs and overestimate performance.[338] There is no reason to assume that transmutation technology, based in part upon breeder reactor technology but involving significant advances in technology and developed by the DOE, will not run into significant cost overruns.
- **Timeframe for Implementation**: The cost estimate was based on a rapid development and deployment scenario which is different than the slower R&D program actually recommended to Congress. As a result, actual deployment would take place later than assumed resulting in a delay in revenue income.[339]

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[333] ATW Roadmap 1999g, p. 2.4
[334] See Fioravanti 1999.
[335] GAO 1996, pp. 3-4
[336] GAO 1996, GAO 1992, NAS-NRC 1999, Gioconda 2000.
[337] See Gioconda 2000, Paine 2000, Fialka 2000, Doyle 2000, and Tri-Valley CAREs 2000 for more information about the cost overruns at NIF. The Department of Energy and the government Accounting Office disagree as to how much of the additional R&D is attributable to NIF. The new costs are now estimated to be between $3.3 billion (DOE estimate) and $3.9 billion (GAO estimate). This is up from $1.2 billion, which was the operative figure at the time that NIF's problems came to light (though even this figure is higher than the originally estimated cost of the facility in 1990).
[338] Merrow et al. 1981 as cited in NAS-NRC 1996, p. 442
[339] ATW Roadmap 1999g, p. 1.1

However, the clearest indication that the costs reported in the Roadmap may be underestimated is the unit reprocessing costs cited in the report. For processing of LWR fuel, the Roadmap estimates a unit cost of $346-406/kgHM.[340] A comparison with the results of the STATS report ($900-$2400 in 1999 dollars) indicates that this could be a very serious underestimate of the reprocessing costs. It is unclear why the STATS panel's revised estimates for reprocessing costs were not taken into account in the Roadmap report. The estimate is approximately six to seven times less than the STATS estimate for an all-aqueous processing facility for private facilities.[341]

Not only is the Roadmap's cost estimate for aqueous UREX processing significantly lower than the aqueous reprocessing costs cited in the STATS report, LWR processing for ATW may be entirely pyroprocessing under one alternative. Unit costs for pyroprocessing are expected to be significantly higher than aqueous reprocessing.[342] This can be seen even within the ATW cost estimate which has a unit reprocessing cost for the ATW fuel (the fuel that has already been in the subcritical reactor and is processed entirely using pyroprocessing). The cost estimate for ATW fuel processing is $5,820 to $7,210/kgHM.[343] The ATW cost estimate also assumes that facilities will be privatized after demonstration. Thus Incorporating a more realistic estimate of front-end reprocessing costs (i.e. using the National Research Council Report's figures) results in a total life cycle cost of nearly $453 billion dollars.[344] This is approximately 1.6 times the undiscounted life cycle cost estimate provided by the Roadmap report. If pyroprocessing

[340] ATW Roadmap 1999g, p. 4.17

[341] The range accounts for the range in unit costs given for different processing units in the ATW cost estimate. However, it does not account for the fact that the ATW Roadmap included decontamination and decommissioning costs (at 10% of construction costs) while the STATS report did not. Thus, the comparison is an underestimate of the relative costs.

[342] Pyroprocessing has often been described as being cost effective and cheaper than traditional aqueous processes. However, it is not always made clear what exactly is being compared. Since pyroprocessing facilities are expected to be much smaller (particularly if each station has its own facility) then the total cost of the facility may be significantly less than an aqueous system. However, the complexity of the process, the high shielding requirements, the lack of economies of scale and the lack of experience in pyroprocessing will make the unit cost high. More recent cost estimates even call into question the assumption that total costs (as opposed to unit costs) will be lower for pyroprocessing. For example the ATW roadmap estimates comparable construction costs (~$500 million) for an LWR fuel processing facility using UREX and processing 175 metric tons per year and an ATW fuel processing facility using pyroprocessing and processing 6.5 metric tons of TRU per year. See ATW Roadmap 1999g, pp. 4.16-4.18.

[343] ATW Roadmap 1999g, p. 4.18

[344] The National Research Council's estimate of privately financed unit costs for aqueous reprocessing was used to adjust the Roadmap's estimate of front-end UREX processing costs. (after adjusting the NAS-NRC 1996 estimate, which was in 1992 dollars, to 1999 dollars). The total cost of UREX processing is estimated by us to be approximately $208 billion as compared to approximately $34 billion in the Roadmap reports. The comparison was done using the unit reprocessing costs and spent fuel throughput for each individual reprocessing facility as provided in the Roadmap report. Using the lower government operation and financing figure of NAS-NRC 1996 would result in a total cost of approximately $80 billion (1999 dollars), still significantly higher than the Roadmap estimate. However, as the Roadmap assumes private financing, the higher figure of $207 billion is more accurate. These figures are higher than those provided by the National Research Council (NAS-NRC 1996, p. 78.) for two reasons. First, because of the adjustment from 1992 to 1999 dollars. Second, because the NRC estimate is based on processing only the 62,000 MTHM of commercial spent fuel slated to be sent to Yucca Mountain while our estimate is based upon the full 87,000 metric tons assumed to be treated in the ATW Roadmap. With these adjustments, the two estimates of total cost for processing LWR spent fuel match.

is chosen as the front-end technology for dealing with LWR fuel, then the costs will be even higher. One estimate is that the unit cost of pyroprocessing LWR fuel would be about 1.57 times the cost of aqueous processing.[345] Using this figure results in a life-cycle cost estimate of $572 billion. Given the fact that the electricity revenues would stay the same, this indicates a significant shortfall, particularly when the discount rate is applied.

This also does not factor in any cost underestimation for other portions of the ATW system (e.g. accelerators and transmuters). For example, the Roadmap estimates a capital cost for the transmutation reactors of $30.2 billion. This means each reactor averages approximately $444 million for 311 kWe or approximately $1430 per kWe. This is approximately 20-25% lower than the cost estimate for the Advanced Liquid Metal Reactor (assuming the same numbers of reactors are deployed).[346] However, this is about half of the construction costs actually incurred by the most recent LWRs to be constructed.[347] Among the factors that have contributed to the increase in costs from earlier, less expensive, LWRs was an increase in the actual costs of construction due to new requirements (e.g. enhanced shielding) and the failure to achieve cost reductions due to learning because of rapid deployment of the LWRs.[348] Both of these factors could very well apply to ATW. There is no reason to believe that the actual construction costs will decrease, as the requirements will stay the same. Also, the proposed program for ATW presented in the Roadmap is one of rapid deployment with little time between reactor construction starts.

The cost estimate also does not factor in the fact that the estimated cost of decontamination and decommissioning provided in the Roadmap (10% of capital costs) is likely to be a severe underestimate, at least for reprocessing facilities.[349] The most recent report of the OECD-NEA which estimates fuel cycle costs uses 30% of capital costs for decommissioning of reprocessing and related fuel cycle facilities for MOX-LWR fuel cycles.[350] This does not account for the fact that ATW fuel cycle facilities will be conducting operations with materials that have higher neutron emissions and overall radioactivity levels than those assumed for either the once-through or MOX LWR fuel cycles.[351] A full consideration of all of the factors listed above (including historical cost escalations) may push the final tab higher than the $453 billion cited above.

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[345] See Gingold et al. 1991 as cited in NAS-NRC 1996, p. 440.

[346] NAS 1995, pp. 318-319. The NAS' estimate of $2,500/kWe for four 303 MWe units was adjusted for inflation to 1999 dollars and then a scaling factor was applied for a cost of $1,850/kWe. The 0.9 scaling factor was applied to estimate the capital costs for 68 modular transmuters of 311 MWe each, following the methodology used by the NAS in Table 6-15, note e.

[347] Bodansky, p. 308 and NAS-NRC 1996, p. 78.

[348] Bodansky 1996, p. 310. On the other hand, the National Academy Panel attributes the cost difference between the 1970s and 1980s largely to changing requirements due to the Three Mile Accident, and very high interest rates in the early and mid eighties, and does not expect that the high costs and wide variations in costs that characterized reactors entering operation in the 1980s to continue. NAS-NRC 1996, p. 78.

[349] The use of 10% of the capital costs comes from a 1978 Nuclear Regulatory Commission report, as cited in ATW Roadmap 1999g, p. 4.11 (note: the text of ATW Roadmap 1999g references NRC 1978 but the reference list cites the report as NRC 1972. The 1978 date is the correct date according to the NRC.)

[350] OECD-NEA 1994, p. 114. The report uses cost estimates from British Nuclear Fuels Limited plc.

[351] ATW would be reprocessing and fabricating new fuel that would contain a higher proportion of the higher actinides since not all reactions, even in a fast spectrum, will result in fission. Furthermore, the length of time scheduled for cooling of the fuel before processing is significantly shorter than in MOX reprocessing operations and thus the short-lived radionuclides have had less time to decay. In fact, the

Given that revenues are highly unlikely to recoup the costs of transmutation and that electricity from ATW will likely not be competitive (even with the likely underestimate of reprocessing costs), the question of ownership of the facilities becomes important. As noted above, the ATW Roadmap calls for privatization after transmuter number two is on-line. There are two possible implications of this. First, the government will pay for research, development and demonstration of ATW. This will amount to $11.147 billion in 1999 dollars. Second, it may not be feasible for subsequent ATW operations to be privatized without significant government subsidies. Unlike current waste management costs, which are paid for out of a fund from fees charged to utilities, ATW costs could be borne directly by taxpayers. This would have serious equity implications. Currently, the fees charged to the utilities are passed on to the ratepayers of those utilities. Thus, the costs are borne by those who receive a direct benefit from the generation of the waste. If the U.S. government subsidizes ATW then all taxpayers will be paying for waste management, including those who did not receive any benefits from the generation of the waste initially. As noted by the National Academy panel, for transmutation to be implemented as a waste management strategy in the United States, "a sustained, long-term national commitment would be necessary. The U.S. government would also have to accept the lead management and financial responsibility, with a cohesive national intent and commitment."[352] This would likely include financial guarantees by the federal government for private industries involved in the transmutation program.[353]

In order for transmutation to have an effect on waste management, it will be necessary to have full-scale implementation of any transmutation scheme chosen. In the case of the US ATW program, for example, this would mean a 118 year campaign with total capital costs on the order of $53.5 billion and average annual operating expenses of $1.8 billion, even before an adjustment to reflect more realistic cost estimating. As noted by the MIT technical review panel, "This is a lot of money to wager on the successful completion of such an extremely complex enterprise, especially when the net gain calculation is based on uncertain economic and technical assumptions."[354]

The MIT review panel goes on to note that this assumes that the facilities operate as planned with 75% availability and 40% thermodynamic efficiency. It also assumes that only 10% of the plant's output would be needed for internal consumption. Even with successful operations, the ATW project may not recoup its costs because the current cost and revenue projections are based on overly optimistic assumptions such as the cost of constructing and operating transmutation systems, the financing costs, the revenues from sales of electricity.

Proliferation

In a 1994 report on plutonium disposition, the National Academy of Sciences noted that "Restricting access to fissile material is the principal technical barrier to

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ability to handle much hotter fuels is one of the stated advantages of pyroprocessing (see for example NAS-NRC 1996, p. 43).

[352] NAS-NRC 1996, p. 8

[353] NAS-NRC 1996, p. 8

[354] Kazimi et al. 1998, p. 5. It should be noted that the Review Panel uses this argument to urge LANL to "make a strong case for the value of spin-offs if the program is terminated before large-scale deployment."

proliferation in today's world, far more so than access to the information and technologies needed to build a weapon once the fissile material has been acquired."[355] Enrichment of natural uranium or separation of plutonium from spent fuel is costly, requires large conspicuous facilities, produces large volumes of waste, and requires some fairly complicated and sophisticated nuclear techniques. Implementation of commercial nuclear fuel cycles that involve reprocessing to separate plutonium has long been recognized as creating proliferation problems. This is one of the major reasons a number of countries (such as the United States and Canada) have a once-through fuel cycle policy.

Implementation of any of the transmutation proposals discussed in this report would result in a significant increase in reprocessing operations and could even result in some countries abandoning the once-through cycle. This would significantly increase the amount of weapons-usable fissile material separated from spent fuel and thereby increase risks of diversion, theft, or abrogation of commitments against proliferation. Weapons-usable fissile materials has been defined by Chow and Solomon of RAND as:

> [U]ranium with a fissile isotopic content of 20 percent or more and plutonium of any isotopic composition. Weapon-usable plutonium includes plutonium separated from the typical spent fuel of commercial nuclear reactors (reactor-grade plutonium) and plutonium from nuclear weapons (weapon-grade plutonium).[356]

There are two important points to be made here. First, there is a difference in the fission behavior of plutonium and uranium depending on the energy of the neutrons. While plutonium-239 and uranium-235 are fissile for any energy neutrons, not all of the plutonium or uranium isotopes are fissile. However, for fast neutrons, as found in a nuclear explosion, all of the plutonium isotopes are fissile and thus any mix of plutonium isotopes is fissile. Second, there is a difference between reactor-grade and weapon-grade plutonium, which is mainly related to the number of spontaneous neutrons generated (higher for reactor-grade), heat generation (higher for reactor-grade) and gamma radiation (higher for reactor-grade). All of these factors make it harder for a weapon designer to use reactor-grade plutonium, both in terms of physically working with the material, but also in designing a weapon with a reliable yield. Thus, the nuclear weapons states produced plutonium in such a way as to minimize the higher plutonium isotopes. But as noted by J. Carson Mark, a former leader of weapons design at Los Alamos National Laboratory, "The difficulties of developing an effective [weapon] design of the most straightforward type are not appreciably greater with reactor-grade plutonium than those to be met for the use of weapon-grade plutonium."[357]

With regard to the differences between weapon-grade and reactor-grade weapons yields, the National Academy of Sciences noted that with reactor-grade plutonium "the probability of achieving only a fizzle yield is several times greater" than for the same design using weapon-grade plutonium, but that "the fizzle yield is not zero."[358] As any mixture of plutonium isotopes will result in a fizzle yield or more, and the fizzle yield of the Trinity test (which was a simple Pu implosion device) would have still yielded about

[355] NAS 1994, p. 26
[356] Chow and Solomon 1993, p. xi.
[357] As cited in NAS 1995, p. 44
[358] NAS 1995, p. 43

1 kiloton, the destructive power would still be devastating. For example, a 1-kiloton fizzle yield would have a destructive radius more than one-third that of the Hiroshima bomb.[359]

The definition of weapon-usable fissile materials must also include radioisotopes that are fissile, but not currently used for nuclear weapons. This includes americium-241 and neptunium-237, which are both fissionable, and both are proposed for separation in some transmutation schemes.[360] It must also include U-233, which is produced from Thorium-232 in systems fueled by a thorium-plutonium fuel (such as the Energy Amplifier). In the case of the Energy Amplifier (as applied to processing Spanish spent fuel), the U-233 is diluted by the uranium from the original spent fuel, which is not completely separated. The resulting product has an enrichment of about 60% U-233. Ignoring the issue of radiation dose due to other radionuclides (addressed elsewhere), this is of a sufficient enrichment for weapons use and results in a critical mass comparable to high-enriched uranium.[361]

Thus, for a party interested in building a nuclear weapon, and which only has access to reactor-grade plutonium, the hurdles are surmountable. In particular, the possibility of a lower yield may not be of concern to all would-be proliferants who might simply be interested in having a weapon, with little regard to the actual yield achieved. It is also interesting to note that reactor grade plutonium may actually pose some advantages to such a group or country. While the higher neutron emissions may result in a lower yield, it also solves two of the major design problems of nuclear weapons: a source of neutrons to start the chain reaction and the timing of the neutron source with the implosion.

Another potential proliferation risk comes from the need for initial loading of fissile materials in some reactors. For example, a critical fast reactor dedicated solely to minor actinide transmutation (i.e. no plutonium fuel) has been proposed as part of transmutation strategy. However, this would create difficulties for safe operation of the reactor so highly enriched uranium (approximately 90% U-235) is also used.[362] This material can also be used for weapons purposes. Similarly, thorium based systems such as the Energy Amplifier or proposed next generation reactors, all require an initial loading of fissile materials since there is no fissile isotope in mined thorium (unlike uranium which contains some U-235). Rubbia et al. in fact propose stockpiling all of the fissile U-233 produced in the Energy Amplifier during the time that transuranics from LWRs are being loaded. Then, when the TRU reduction mission is complete it would be possible to continue the reactors with the stockpile U-233.[363] Of course, during the intervening years, a significant stockpile of weapons usable material would accumulate.

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[359] NAS 1995, p. 43-44

[360] See Albright and O'Neill, eds. 1999 for more information on Am-241 and Np-237.

[361] Pistner 1999, p. 63

[362] OECD-NEA 1999b, p. 163-164. In this particular example, the fuel consists of approximately 65 percent minor actinides (Np, Am, Cm) and 35 percent HEU. Both a smaller number of delayed neutrons (necessary for reactor control) and a reduction in the effectiveness of reactivity feedback mechanisms are the cause of safety concerns in a critical fast reactor fueled solely with minor actinides.

[363] Rubbia et al. 1997b, pp. 52-53

Though the distinction is a false one set up by proponents of pyroprocessing, the following two sections shall discuss the proliferation implications of transmutation in terms of both the expansion of conventional PUREX-based reprocessing and in terms of the introduction of new reprocessing techniques which do not result in separated plutonium.

Potential for Expansion of PUREX

Implementation of transmutation programs would likely result in a major expansion of PUREX-based reprocessing operations. Unlike the United States, which currently does not undertake any commercial reprocessing and which is focused on accelerator transmutation of waste (more on this below), there are other major commercial nuclear powers which do use PUREX. Furthermore, their transmutation proposals rely upon a combination of light water reactors, fast reactors, and possibly accelerator based systems. These proposals would use PUREX and PUREX based reprocessing operations (with the possible addition of pyroprocessing for some of the fuel either in the fast reactors and/or the accelerator based systems). Implementation of transmutation in countries such as France, the UK and Japan, would likely lead to pressure in favor of transmutation in other countries using nuclear power (e.g. South Korea, China, Taiwan) and a subsequent further expansion of PUREX processing. The net result would be a massive increase in the amount of separated plutonium. The amount of commercial plutonium (both separated and in spent fuel) is far greater than the military stocks of the nuclear weapons states. Separation of this plutonium would vastly increase proliferation risks.

As one quarter of all operating nuclear power plants are located in the United States, the future of US reprocessing plans is important. Currently, in order to minimize the proliferation risks of commercial nuclear power, the United States does not engage in reprocessing of commercial plutonium. This is explained in a September 1993 White House Fact Sheet on Non-Proliferation and Export Control Policy which states, that "The United States does not encourage the civil use of plutonium and, accordingly, does not itself engage in plutonium reprocessing for either nuclear power or nuclear explosive purposes."[364] However, Congress, in both 1999 and 2000, attempted to pass a nuclear waste bill which would have included the establishment of an Office of Spent Nuclear Fuel Research. Among other tasks, this office would "require research on both reactor- and accelerator-based transmutation systems" and "require research on advanced processing and separations." The goal is to "investigate technologies for the treatment, recycling, and disposal of spent nuclear fuel and high-level radioactive waste."[365] The President vetoed these bills, but more recently, the provision was included in a new bill called the National Energy Security Act of 2000, which is still in the Senate.[366]

Furthermore, the next step in the ATW Roadmapping effort is a series of trade studies to determine the best mix of technologies to pursue. It will not be limited to

[364] This White House Fact Sheet (White House 1993) is widely believed to be a public version of official non-proliferation policy adopted at the same time and codified in Presidential Decision Directive 13.
[365] S.1287 Nuclear Waste Policy Amendments Act of 2000, Sec. 302.
[366] S.2557 The National Energy Security Act of 2000, Sec. 416.

accelerator based systems and but would also examine critical reactor systems, which would by necessity require aqueous PUREX reprocessing.[367] Efforts to bring back PUREX processing in the United States are not new, however. For example, in a 1995 letter from Senator Strom Thurmond to Senator Frank Murkowski, the Senator from South Carolina (the location of one of the United States military reprocessing facilities) recommends the formulation of a nuclear waste plan that includes "at minimum" the "Construction and funding of storage and reprocessing facilities at SRS specifically for commercial, research (foreign and domestic) and other DOE spent fuel, along with legislative mandates that reprocessing, once begun, not be interrupted."[368] These efforts may be redoubled if transmutation is implemented overseas using PUREX or other aqueous systems. The US, if it focuses only on ATW, could end up lagging because of the longer development timeframe for an ATW only system.

A decision by the United States to overturn its policy on reprocessing would have severe proliferation consequences. First, it would result in significantly more separated plutonium. Second, the United States would no longer be able to lead by example in discouraging others to avoid reprocessing in its efforts to combat proliferation. Third, as the United States has effective veto power over whether overseas spent fuel made with US uranium is reprocessed, a resumption of US reprocessing would weaken its position that those countries should not reprocess their spent fuel.

Transmutation based on PUREX could also significantly increase the risk over PUREX solely for MOX purposes. In addition to the separation of plutonium, it would also involve the separation of americium and possibly neptunium for transmutation. Both Am-241 and Np-237 are weapons-usable fissile materials, as discussed above. Their separation would thereby increase the overall amount of separated fissile materials available for weapons use.

New Reprocessing Techniques

There are a number of new reprocessing techniques being developed that have application to nuclear waste transmutation proposals. While some involve separation of plutonium, others separate out the transuranic elements as a group. One such group of processes is based on electro-metallurgical techniques (which often go under the term pyroprocessing).

Proponents of both fast breeder and some accelerator based systems make the claim that the electro-metallurgical processing likely to be used would have a much higher degree of proliferation resistance. This is because pyroprocessing separates the transuranic elements as a group. While PUREX results in a product that is nearly 100% plutonium, pyroprocessing results in a product that is a mixture of plutonium, americium, curium and neptunium. While it is true that pyroprocessing, unlike PUREX, does not produce pure "naked" plutonium, the purported proliferation resistance has been overstated. This overstatement can be seen in the following quote from a LANL document:

[367] Personal notes of Hisham Zerriffi at the December 6, 1999 meeting of the Department of Energy, Office of Nuclear Energy, Nuclear Energy Research Advisory Council (NERAC).
[368] Thurmond 1995.

> In the ATW concept, spent fuel would be shipped to a ATW site where the plutonium, other transuranics and selected long-lived fission products would be destroyed by fission or transmutation in their only pass through the facility. This approach contrasts with the present-day reprocessing practices in Europe and Japan, during which high purity plutonium is produced and used in the fabrication of fresh mixed-oxide fuel (MOX) that is shipped off-site for use in light water reactors. Instead of "reprocessing," the ATW approach can be fairly characterized as "once-through destruction." ATW would inhibit plutonium accumulation, proliferation and diversion.[369]

Contrary to the claim, plutonium is not destroyed in its "only pass through the facility." If this were the case, the backend processing system would only have to separate out long-lived fission products from short-lived fission products. In fact, there is only about a twenty or thirty percent burn-up of the transuranic elements in an accelerator based system. As a result, there would be about four metric tons of transuranic material separated in each pyroprocessing facility per year.[370] This means a total of 32 metric tons of transuranic materials will be separated per year when all eight facilities are running. This is in addition to another 15 metric tons of transuranic material per year separated from the initial light water reactor fuel. Thus, in any given year there would be up to 47 metric tons of separated weapons-usable materials.[371] Thus, while the processing and transmutation facilities may be co-located, it does not follow that these facilities can be fairly characterized as "once-through destruction."

Furthermore, while it is true that the pyroprocessing system does not produce pure "naked" plutonium, it does separate the transuranics. As noted by the MIT review, "removing Pu from a material composed of Np, Pu, Am and Cm is in principle much easier than separating Pu from spent uranium fuel. Therefore, the proposed ATW fuel might not meet the current spent fuel standard for proliferation resistance. Further consideration of this point is needed to assess its implications for the required controls for the deployment of the ATW fuel system."[372]

The greater ease of plutonium separation from the pyroprocessing product as compared to spent fuel is fairly easy to understand. Separation of PU from spent fuel involves handling large quantities of material (the uranium makes up approximately 94% of the light water reactor spent fuel). It also involves handling material that contains highly radioactive fission products. However, if the uranium and fission products have already been largely separated as part of a "waste management" process, these problems are reduced. Further processing of the transuranics could be done on a very small scale (possibly even in a glovebox) and the shielding requirements would be lessened.[373] This processing could be the result of modification of the pyroprocessing cells (the chambers in which pyroprocessing takes place) or implementation of small-scale PUREX based

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[369] Venneri et al. 1998, p. 2

[370] ATW Roadmap 1999d, p. 38.

[371] It should be noted that this full amount of separated transuranics would only exist in those years that all eight stations are operational. In the early and late years, this amount will be less, as not all eight facilities will be on-line at once.

[372] Kazimi et al. 1998, p. 4

[373] The transuranics include some gamma emitters that would have to be taken into account. Also, there is a small amount of lanthanide fission products that remain with the actinides. These fission products include gamma emitters. Thus, shielding requirements would still be stringent, but less so than when processing spent fuel which contains higher amounts of fission products.

processing. Such a facility would be significantly smaller than a conventional PUREX facility and the materials being processed would not contain volatile radionuclides, whose emissions (particularly to the air) can be monitored by international inspectors. A covert facility of this type would likely be more difficult to detect.

However, it may not be necessary to have complete separation of the plutonium in order to have weapons-usable fissile materials. Two of the minor actinides are fissile with both thermal and fast neutrons (Np-237 and Am-241) and can sustain a chain reactor on their own, while the others will fission with fast neutrons (as occur in nuclear weapons). Therefore, direct use of a pure transuranic mixture may be limited more by materials issues (e.g. heat and gamma emissions) than by the plutonium content of the fissile materials.

The proliferation risks of pyroprocessing were previously examined in a report by Martin Marietta for the Departments of State and Energy.[374] This was done in the context of the Integral Fast Reactor (IFR) program, an Advanced Liquid Metal Reactor (ALMR) being developed for the breeder reactor program that would have used pyroprocessing. Much of that program is now being repeated in the ATW program in the United States and the conclusions are still relevant. One significant difference between the IFR program and the ATW program would be the use of non-fertile fuels in ATW (i.e. the fuel would not use uranium). The presence of uranium was identified in the IFR report as one of the barriers to the use of pyroprocessed transuranics in a nuclear weapon. In the ALMR/IFR system, the pyroprocessing product would have been approximately 70% plutonium and 30% uranium.[375] However, implementation of pyroprocessing for transmutation would result in a product that is approximately 99.9% transuranic actinides.[376]

A more recent report evaluated the proliferation risks of electro-metallurgical treatment for DOE sodium bonded spent fuel (this would have used a subset of the pyroprocessing technology) and was largely based on the earlier report by written by Wymer et al. for Martin Marietta.[377] The Wymer and DOE reports found a number of potential proliferation risks in pyroprocessing such as:

- Pyroprocessing requires bulk handling of plutonium which makes plutonium accounting more difficult.[378]
- IAEA safeguards have not been established for this type of processing. Furthermore, the plutonium accounting method developed by Argonne National Laboratory (ANL), the developer of the process, for the IFR project is likely to be inadequate.[379]
- Adequate sampling for materials accounting will have larger uncertainties since "*representative sample collections from the IFR fuel recycle process are difficult because of the inherent nature of the process*."[380] (emphasis in original)

[374] Wymer et al. 1992
[375] OTA 1994, p. 20-21
[376] ATW Roadmap 1999d, p. 6
[377] DOE 1999a
[378] Wymer et al. 1992, p. 58
[379] Wymer et al. 1992, p.58

- Accounting will be made more difficult because of large holdup of materials. One particular area of concern is the electrolytic cell where special nuclear materials (SNM) inventories "can fluctuate significantly during batch operations."[381]

Both the Wymer report and the DOE report conclude that the proliferation risks of pyroprocessing are manageable. In part this is due to an assumption that strict export controls would be placed on the technology and in part due to the fact that pyroprocessing does not separate plutonium and that there is still a radiation barrier. One conclusion of the Wymer report is that any nation with a serious intent to acquire fissile materials is unlikely to opt for developing pyroprocessing. Rather the better known and technically easier PUREX process would be the logical choice. However, if pyroprocessing is widely implemented, particularly as a waste management technology, then it may be seen as a more likely candidate. Its use would be legitimized and probably promoted widely. The Wymer report notes that if IFR were to be implemented for the purpose of reducing actinide inventories (i.e. for waste management) rather than as a breeder, implementing export controls may be more difficult. "If the U.S. expresses the view that it is primarily interested in IFR as a potential tool for reducing the inventory of actinides in spent fuel from LWRs, then, conceivably, any nation with a significant investment in light water reactors can express a legitimate interest in having a close involvement or access to the IFR program."[382] Of course, the United States is not the only country developing pyroprocessing techniques. The issue of controlling the spread of pyroprocessing technology is discussed in the section below titled "Technology Transfer."

It is also not clear that pyroprocessing will be able to be implemented as a complete technology for processing of light water reactor fuel. The recent ATW Roadmap leaves open the possibility of implementing pyroprocessing for both LWR fuel and for ATW fuel. However, the baseline considered in the report for LWR fuel is a combination of aqueous processing based on PUREX and pyroprocessing. Incoming spent fuel from reactors would go through what is being called the UREX process. UREX is a solvent exchange process very similar to PUREX and would be used to separate the uranium, technetium, and iodine. This would also release some of the gaseous fission products. Only then would the remaining material, consisting of the actinides and the remaining fission products undergo an electro-metallurgical treatment.

This significantly increases the proliferation risk of implementing transmutation. Using a PUREX-based process on the front-end creates two new risks not present in the baseline pyroprocessing technology. First, if the technology is disseminated for waste management purposes it would significantly spread the knowledge of PUREX processing and provide experience in aqueous reprocessing. Second, it provides a new area and method for diversion. Modification of the UREX process or diversion of the aqueous stream containing the plutonium to a significantly smaller PUREX processing facility are two new possible methods for diversion. This is particularly problematic since the materials would be put into a form most amenable to efficient plutonium separation and the plutonium isotopic composition would be at its most ideal from a weapons production

[380] Wymer et al. 1992, p. 62
[381] Wymer et al. 1992, p. 64
[382] Wymer et al. 1992, p. 74

standpoint. As noted above, the isotopic composition of the plutonium and the presence of minor actinides, is not necessarily a deterrent to the use of materials for weapons purposes. However, separation of plutonium from incoming LWR fuel, as opposed to separation from fuel that has been irradiated in the transmuter, would result in a product which minimizes some of the handling problems associated with the higher plutonium isotopes and minor actinides.

Modification of the Reactor Core

Another factor which must be considered in assessing the potential for transmutation technologies to aid in proliferation is the possible modification of reactors in order to produce greater quantities of, or isotopically purer, plutonium. There are a variety of ways in which this could be done. For example, the National Research Council report on transmutation noted that:

> In addition to fuel rods in the reactor core that contain plutonium, a fast reactor has blanket regions for breeding ^{239}Pu in rods that contain ^{238}U (natural or depleted uranium) and that could be changed while the reactor is operating (on-line). The blanket would be a special target in diversion scenarios that include covert substitution of blanket rods, which are taken away and reprocessed to recover the ^{239}Pu.[383]

In reactor configurations such as those being proposed for the U.S. ATW program, the central core consists of a non-fertile plutonium fuel in a fast neutron flux. However, the long-lived fission products would be located in targets external to this core ("ex-core targets") and the neutrons would be thermalized in order to increase the absorption cross-section. These ex-core regions function much like the breeding blankets of fast breeder reactors. Replacement of these Tc-99 and I-129 targets with suitable targets made from U-238 (which would be available from the processing of LWR fuel) would result in plutonium production in the targets. Processing of such targets to remove the plutonium could be done in small-scale facilities. There are a number of questions about the feasibility of such a scheme that must be addressed. For example:

1. How would ex-core production of plutonium affect reactivity and safety of the reactor (since some of the plutonium would fission in a region not originally designed for fission reactions of plutonium)?
2. How will safeguards for transmutation systems address ex-core transmutation targets? Current safeguards are focussed on the reactor fuel and have not had to address this issue in the past. The feasibility of safeguarding ex-core targets would have to be addressed.
3. What size and type of reprocessing facility would be most suitable for ex-core U-238 targets (given the assumption that reprocessing is being conducted for covert separation of plutonium)?
4. How much plutonium could be produced in such target and how long would it take to separate a significant quantity of plutonium?

[383] NAS-NRC 1996, p. 373

Technology Transfer

As noted in the above section, control of the technology to separate fissile materials plays a large role in determining the proliferation impact of these technologies. This relies on a fundamental assumption: it is possible to prevent and control the dissemination of technology. In order to address this question, we will briefly examine two modes for the transfer of technology: purposeful technology transfer and inadvertent technology dissemination.

In some cases, the development of technology in one country leads to the purposeful transfer of that technology to other countries. President Eisenhower's Atoms for Peace program was a perfect example. Today, proponents of reprocessing technologies for transmutation propose reliance on export controls and similar regulations to ensure that technologies would not be transferred (or would be transferred in a limited manner). However, it may be very difficult to restrict sales (for example, to only countries currently reprocessing using PUREX). Countries relying on nuclear power could have a legitimate argument for purchasing these technologies, even if they do not currently reprocess their spent fuel. After all, if reprocessing and transmutation were a legitimate waste management technique it would be hard to deny such benefits to others in a similar situation. In fact, Article IV of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) calls for such nuclear cooperation.

Even without purposeful dissemination of technologies (nuclear or otherwise), there is a certain inevitability that once a technology is developed in one country, other countries will obtain the necessary skills and knowledge to develop the technology if they desire. There are a number of examples of failure in the control of sensitive technology, ranging from the proliferation of nuclear weapons to development of missiles to sales of supercomputers. Nuclear weapons are one perfect example, with eight countries currently believed to be in possession of nuclear weapons, others suspected of having nuclear weapons programs and a number of countries with the technical knowledge and skills to develop nuclear weapons should they desire (e.g. Germany or Japan). Therefore, there is no reason to leap to the assumption that new reprocessing technologies would not be disseminated, particularly when the historical record indicates that prudence and vigilance are necessary.

Given the above arguments concerning the proliferation risks of both PUREX and new reprocessing techniques and the discussion of technology transfer, are there any reasons to encourage the spread of reprocessing? The argument has been made that separation and transmutation of plutonium actually reduces the risk of plutonium proliferation. The argument is that any repository is, in effect, a "plutonium mine." There is only one sure way of reducing the proliferation dangers of plutonium and that is to get rid of it.[384] This argument is the subject of an article by Dr. Edwin S. Lyman of the Nuclear Control Institute and Dr. Harold A. Feiveson of Princeton, both experts on non-proliferation and nuclear fuel cycles.[385] The article concludes that operations to remove spent fuel from a repository with the intent to reprocess it and obtain weapons-usable

[384] See for example, ATW Roadmap 1999a, p. 5-4 which states that "The most complete method for ensuring that fissile material is never used for nuclear explosives is to transmute it into something that is not fissile." Bowman 1997, p. 135 also notes the possibility of repositories becoming mines for plutonium.
[385] Lyman and Feiveson 1998.

materials would be an extremely costly and lengthy process that would be easily detected. Furthermore, any country with an existing nuclear infrastructure can more readily separate plutonium from its operating facilities than mine it from a repository. As for the transmutation alternative, the authors note that any such endeavor would require a large number of facilities, would result in separation of transuranic materials, and would take centuries. In addition, safeguarding the array of facilities required for transmutation (which include not only reprocessing facilities, but also fuel fabrication and waste processing facilities) would be more difficult than safeguarding a repository. Not only would there be fewer facilities to safeguard in the repository scenario, the material would be in a form more amenable to stringent safeguards (single items, spent fuel casks, rather than bulk amounts of plutonium and transuranics which require more difficult materials accounting procedures).[386]

In addition to the particular points made by Lyman and Feiveson, the argument of "plutonium mines" being more dangerous than reprocessing and transmutation can be taken to two logical conclusions, neither of which bode well for reducing nuclear dangers (nor necessarily make common sense despite their logic). The first logical conclusion is that reprocessing technology should be disseminated on a limited scale to only those countries that are not expected to pose a proliferation risk. This would significantly reduce plutonium stockpiles worldwide while still not transferring the technology to certain countries. However, at the end of this process most of the plutonium stocks remaining would be in the hands of those considered to be a proliferation risk.

The second logical conclusion is that eliminating the risk of "plutonium mines" would necessarily require the purposeful dissemination of reprocessing technology worldwide to every country with nuclear reactors. If all repositories are potential "plutonium mines" then only full-scale implementation of transmutation will serve as a solution. Therefore, all spent fuel worldwide would have to be processed, including that in countries suspected of having nuclear weapons programs.

In essence, the problem is one of having your cake and eating it too. Proponents of transmutation cannot advocate it as solving the problem of potential plutonium separation in the future if their solution results in widespread plutonium separation now.

The overall conclusion to be drawn regarding the proliferation consequences of widespread implementation of reprocessing and transmutation is that the risks would increase. This was also the conclusion of the National Research Council panel, which stated:

> Nuclear proliferation is an issue even for the once-through fuel cycle, where it is addressed by domestic security measures and especially by international safeguards to deter the misuse of reactors, enrichment facilities, and stored spent fuel. Proliferation risks would generally be greater with widespread implementation of S&T [Separations and Transmutation] systems in the many nations using nuclear power, mainly because of two factors: (1) the availability of bulk quantities of plutonium in separated or readily converted form at various places in the fuel cycle, which can be a challenge for safeguards even with stringent materials accountability and surveillance systems; and (2) the

[386] See Lyman and Feiveson 1998, pp. 126-127.

availability of large reprocessing facilities that could be misused for production of fissionable weapon materials, e.g. after treaty withdrawal or abrogation. The Clinton administration's policy announced in September 1993 reaffirms the link between U.S. nonproliferation goals and concerns vis-à-vis civil plutonium reprocessing and its use in nuclear power, as emphasized by the United States in the late 1970s, and discourages any S&T undertaking with LWR spent fuel in the United States for the foreseeable future.[387]

Environment, Safety and Health

The civilian nuclear fuel cycle as well as the operation of nuclear facilities for military purposes has already resulted in a legacy of waste and environmental contamination which will pose a threat to the environment, safety and health for generations to come. The question is what new ES&H implications could arise from implementation of transmutation.

Spallation and Neutron Activation Products

It is already widely known that the large neutron flux of nuclear reactors creates a problem in the creation of large amounts of "neutron activation products." Stable isotopes (for example in the cladding of the fuel, the reactor vessel or piping) can absorb a neutron and become radioactive. This raises the cost of nuclear facilities since they require decontamination and decommissioning at the end of their use. This also creates another source for large volumes of waste. Any transmutation scheme would, of course, involve a high level of neutrons and therefore D&D will be an issue with any of the programs.

Accelerator based systems face some unique challenges however. First, the neutron flux in accelerator systems is over ten times higher than in commercial thermal reactor systems.[388] This high neutron flux could increase difficulties in disposing of neutron activated waste, such as the reactor components themselves during decontamination and decommissioning. Neutron activation of zirconium would be a particular problem. Zirconium would form a large part of the fuel in some ATW systems, replacing the major component of ordinary reactor fuels, the fertile uranium. This would likely result in significant production of Zr-93, a long-lived radionuclide (half-life 1.53 million years) which is considered an important contributor to the dose under some repository conditions.

Second, in addition to neutron activation products in the reactor, accelerator based systems must deal with radioactive products in the spallation target. These are produced by the interaction of the protons and the spallation source used to produce neutrons (spallation products) and by neutron activation of the spallation target. The residual isotopes left after neutron production vary widely and are often radioactive. One such product is polonium-210 from neutron capture by bismuth (in those systems that use lead-bismuth eutectic as a target as well as coolant). Po-210 is an alpha-emitter and has a

[387] NAS-NRC 1996, p. 108

[388] NAS-NRC 1996, p. 28. Though this comparison was to a different ATW system than is currently proposed, the two ATW systems have similar flux levels.

half-life of 138 days and thus can pose a concern for operation of the facility due to its high production and short half-life. In particular, contact of the lead-bismuth eutectic with air (for example, if there is a coolant leak) can create polonium bearing aerosols and volatilization of the polonium in the form of a polonium hydride.[389] Otherwise, the polonium radioactivity is mitigated by the fact that the Po-210 stays in the lead-bismuth eutectic and the lead acts as a shield against the radioactivity of the Po-210.

However, Po-210 is not the only radionuclide potentially produced in the spallation target that can pose problems. Target experiments in Russia[390] resulted in a variety of radionuclides being produced such as mercury, cesium, xenon, and bromine (various radioactive isotopes of each).[391] After three months of irradiation, the total activity of the target was over 1000 Ci/kg.[392]

Operational Worker and Population Doses

The separation of the spent LWR fuel and liquid high-level waste into different product streams could have a serious effect on worker (and potentially public) radiation doses. In addition to the increased risk that can be expected from a major expansion of nuclear chemical operations due to implementation of S&T there are a few specifics of S&T which are cause for concern.

Pyroprocessing of spent fuel would involve processing of the highly radioactive fuel under conditions not experienced before. The volumes are much smaller and there is a much higher concentration of radioactive materials. For example, the MIT review notes that the small volumes used in processing spent ATW fuel "will have extremely large radiation doses."[393] In addition to the effects on the chemistry of the process and the equipment, noted by the MIT Review Panel, there would also presumably be an effect on workers that must be addressed. This is compounded by the fact that pyroprocessing cooling times will be shorter than conventional reprocessing.[394]

Another example of the added potential for worker and population doses comes from the need to fabricate fuels containing high gamma emitting minor actinides (such as americium and curium). The potential dose from these radionuclides establishes certain technology limitations on transmutation proposals. In particular, the shielding requirements to protect workers from gamma emissions of both the americium and the gamma-emitting lanthanide impurities severely limits the amount of americium which can be contained in uranium oxide fuels. This necessitates the use of inert matrix fuels fabricated in specially designed process facilities.[395] However, americium may not

[389] Gromov et al. 1997, p. 208.
[390] The target was 20cm in diameter, 60cm in length, and was subjected to an 800 MeV, 1.25 mA proton beam at two different irradiation intervals. See Shubin et al. 1999, p. 1.
[391] Shubin et al. 1999, p. 4-5
[392] Shubin et al. 1999, Figure 1, p. 2
[393] Kazimi et al. 1998, p. 4
[394] OECD-NEA 1999b, p. 36. The shorter cooling times are possible because of lower sensitivity of these processes to radiation damage and criticality constraints.
[395] OECD-NEA 1999b, p. 38.

perform well in all such fuels.[396] Curium poses even greater difficulties, and some transmutation proposals may require storage of curium for 100 years.[397] This would preclude some transmutation scenarios. In any case, processing these materials increases the risks of both routine and accidental exposures. In particular the potential source-terms, if the containment of the materials fails, are much higher with these fuels.[398]

Finally, another potential new dose risk would be the separation and storage of the short-lived fission products. The high concentrations of these short lived fission products, now separated from the much larger volume of Uranium fuel and zirconium cladding, would have to be assessed for their potential dose contributions to workers and the public. This is, of course, complicated by the fact that if storage is chosen as the option for handling the medium lived fission products, these radionuclides will be above-ground in human-engineered facilities for around 300 years or more.

Nuclear Fuel Cycles

Implementing a full-scale separations and transmutation program would be a significant change in the nuclear fuel cycle of any country. In some cases (e.g. the United States, Sweden, Canada) it would mean a complete abandonment of the once-through cycle. For others (e.g. the U.K., France, Japan) it would mean a significant expansion of reprocessing. For all of these countries it would result in a significant expansion of their nuclear infrastructure, requiring new processing and waste management facilities and a number of new reactors. Beyond a change in fuel cycle, implementation of transmutation would have significant impact on the future of nuclear power.

Many of the proponents of transmutation of LWR fuel see it as a transition step to a new fuel cycle.[399] The possible fuel cycles being considered are varied and range from wider implementation of MOX fuel in LWRs to completely accelerator-based nuclear power systems breeding new fissile material from thorium. Figure 21 shows some examples of new fuel cycles that would use transmutation.

Various combinations of the different types of reactors represented in Figure 21 can be included in a particular transmutation scheme. However, certain limitations in reactor performance require the use of certain reactor types. Use of plutonium in MOX fueled light water reactors is limited in its applicability and has generally been limited to one pass through the reactor (this is due to the build-up of the minor actinides and their effect on core reactivity and fuel fabrication). In order to utilize the plutonium further or to fission the minor actinides a fast burner would be required. However, the criticality requirements for fast critical reactors limit their effectiveness in reducing the inventory of actinides. Thus, accelerator based systems become necessary for further reduction of actinide inventories.

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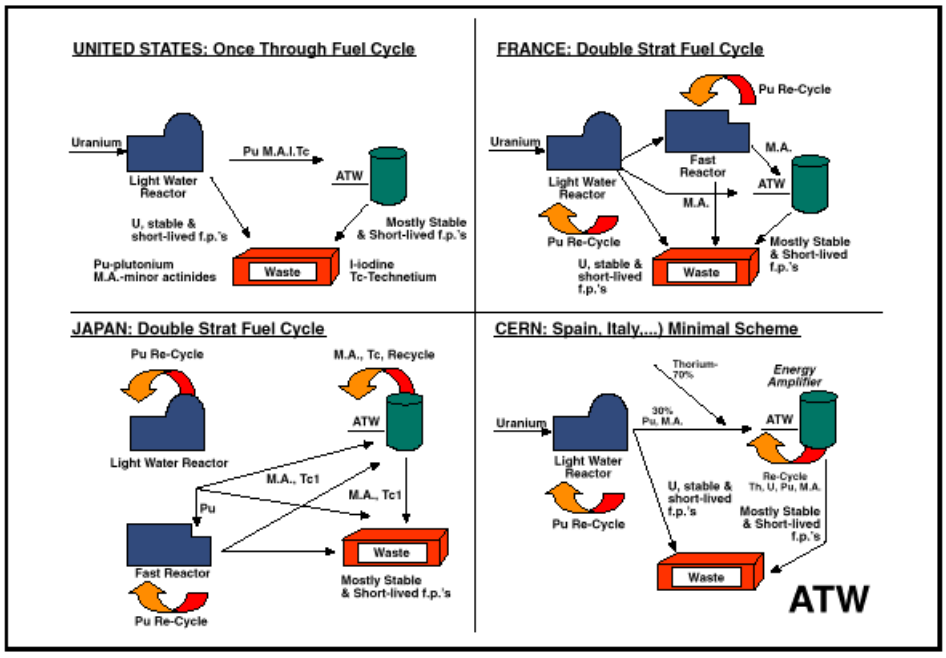
[396] OECD-NEA 1999b, p. 38. In particular, some of the properties of americium oxide make it difficult to use in inert fuels based on oxides.
[397] OECD-NEA 1999b, p. 38 and Salvatores and Zaetta 1997, p. 113.
[398] "Source term" refers to the amount of radioactivity released. In addition to actual release amounts, it can also refer to potential release amounts.
[399] For example, Bowman 1997, pp. 135-136 and Venneri et al.1998, p. 3

Figure 21: New Fuel Cycles



Source: Figure 3.1 of ATW Roadmap 1999a

Such fuel cycles would involve significant reprocessing, fuel fabrication, and reactor operations to implement. These operations would be made more difficult by the presence of the highly radioactive minor actinides as discussed above. The advantage of such a reactor mix would be a decrease in uranium requirements with its attendant health and environmental risks due to mining, milling, and waste disposal.

New Thorium Based Fuel Cycles

For decades, various programs have been in place to develop a nuclear fuel cycle based upon Thorium-Uranium. Neutron capture on thorium-232 would produce U-233 by the following reaction (with half-lives of intermediate radionuclides shown in minutes and days):

Th-232 + n → Th-233 (22.3m) → Pa-233 (27d) + e → U-233 + e

U-233 is a fissile isotope of uranium, meaning that it fissions with low energy neutrons and can sustain a chain reaction (see Appendix A). As stated in a report by the International Atomic Energy Agency, there are two purported advantages of the Th-U cycle:

1) The Thorium-Uranium cycle produces a relatively small amount of higher actinides compare[d] with Uranium-Plutonium cycle, because of the small capture to fission ratio in ^{233}U and because of the presence of two other fissionable isotopes of Uranium (^{235}U and ^{237}U) in the chain leading to Plutonium and the other heavier actinides[400]

[400] As explained in Chapter I, when a nucleus of a fissionable isotope interacts with a neutron it can either fission or it can capture the neutron to form the next heaviest isotope of that element. The capture to fission

2) The Thorium-Uranium cycle is regarded as safer than the Uranium-Plutonium cycle from a nuclear weapons proliferation standpoint, because of the presence of the hard-gamma emitter in the ^{232}U decay chain as a minor product of the cycle, and because of the possibility of straightforward isotopic dilution of ^{233}U with depleted or natural Uranium in the feed or start-up fuel.[401]

While both of these statements are true, these two "advantages" cannot be achieved simultaneously. In order to realize the first advantage (avoiding the production of actinides) it is necessary that there be no U-238 in the fuel since it is neutron capture by U-238 that forms plutonium (and then the higher actinides when the plutonium captures a neutron). But, the second advantage (the dilution of the U-233, can only be realized if U-238 (the major component of depleted and natural uranium) is intentionally added to the fuel, thus negating advantage number one.

One of the stated advantages of a TH-U accelerator-based nuclear energy cycle is the increased proliferation resistance as compared to the U-Pu cycle. This stated proliferation advantage is due to the following reasons:

- Thermal spectrum Th-U systems have less weapons-usable material as compared to the fast spectrum used for U-Pu breeder reactors.[402]
- The presence of U-232, which has decay products that emit high-energy gamma radiation (the most notable of which is thallium-208), increases the shielding requirements and handling difficulties of U-233.[403]
- U-233 can be diluted with U-238 to make it non-weapons usable.[404]
- Minimal formation of actinides, including plutonium.[405]

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ratio is a measure of whether nuclei is more likely to capture the neutron or to fission (a capture to fission ratio of exactly one means either is just as likely, a ratio less than one means fission is more likely, and a ratio more than one means capture is more likely). The fact that U-233 has a small capture to fission ratio means it is more likely to fission than to be captured. It should be noted that this does not measure the absolute probability of either occurring, only their relative probabilities. As for the isotopes between U-233 and the higher actinides, to be accurate, the isotopes identified are "fissile" as opposed to simply being fissionable. That is, they can be fissioned by low energy neutrons.

[401] IAEA 1997a, p. 15. A hard gamma-emitter is one that emits an energetic gamma ray. The high gamma emissions make handling of the U-233 more difficult.

[402] Bowman 1997, p. 148. While this may be true for critical reactors, it is not clear that this advantage remains significant with accelerator based systems. In a critical breeder reactor, a fast spectrum is often used because it produces more neutrons per fission that can be used to produce fissile materials. However, at the same time, in a fast spectrum the cross-section for plutonium is lower than in a thermal spectrum and therefore more plutonium is required to have enough fissions to keep the reactor critical. In an accelerator based system, the accelerator acts as a supplemental source of neutrons no matter whether the spectrum is thermal or fast or the fuel is Th-U or U-Pu. These supplemental neutrons could be used for both breeding and fission. Thus, an accelerator based U-Pu breeder may be able to use a thermal spectrum and a lower amount of fissile material. It is necessary to make sure that apples are being compared to apples and not to oranges as would be the case if an accelerator based Th-U system is compared to a critical U-Pu system, as it appears that Bowman has done.

[403] Wilson 1999, p. 3

[404] Wilson 1999, p. 3. It is noted in this paper that there has been concern expressed with using uranium facilities for thorium operations (which the author notes is likely due to thorium's greater radiotoxicity and may not apply to uranium use in thorium facilities).

In some cases, the proliferation risk reduction results in an increase in risk in another area. For example, the build-up of U-232 in irradiated thorium fuel results in significant gamma emissions. While this would make handling of U-233 difficult for weapons production purposes, it would also make U-233 handling difficult for fuel fabrication purposes. Thus, a Th-U fuel cycle would have increased costs and complexities due to the shielding requirements and potentially more serious worker risks.

Thorium-uranium fuel cycles also need a start-up fuel since thorium does not have any fissile isotopes (unlike uranium). Thus initial fuel fabrication for the reactor will involve handling separated plutonium or uranium. If the uranium were LEU, then this would include U-238 and therefore the production of actinides, one of the results this fuel cycle seeks to avoid.[406] If the uranium is HEU, then there is the risk of diversion of the start-up fuel. The dilution of U-233 with U-238 would pose the same problem as using LEU as a start-up fuel.

The thorium-uranium fuel cycle would also carry many of the other risks associated with the U-Pu fuel cycle. Reprocessing of thorium-uranium fuels would be costly, create various wastes, and pose health risk to workers and the public. Reactors fueled with Th-U would still have the safety problems associated with all nuclear reactors.

Expansion of Nuclear Power

The prospect of transmuting what are considered to be the most problematic radionuclides in spent fuel is inextricably linked to a continuation and expansion of nuclear power. By "solving" the nuclear waste problem, proponents believe that nuclear power would become politically acceptable again. Proponents of ATW also point to the safety advantages of a sub-critical system and the self-contained operation of the facility (i.e. collocated reactor and reprocessing facility) to minimize proliferation risks. This would be the hat-trick of the nuclear industry, solving all three of the major problems with nuclear power.

Transmutation has been discussed in the context of a phase-out of nuclear power, level continuation of current nuclear power usage, and expansion of nuclear power. However it is clear that the proponents of transmutation, who come mainly from within the nuclear industry, view transmutation as a key link in securing the future of nuclear power. The Foreword to an IAEA Status report on accelerator transmutation of waste begins:

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[405] Wilson 1999, p. 4. Wilson does note that this advantage will be minor, unless current stocks of minor actinides, particularly neptunium-237, are reduced through reprocessing and transmutation.

[406] Bowman 1997, pp. 148-149. Bowman also raises the possibility of starting with no initial fissile material in the reactor. In this case, the accelerator would use electricity from the electric grid to produce spallation neutrons that would produce U-233 from the thorium in the reactor core. These U-233 nuclei would fission producing more neutrons and therefore more U-233 from the thorium. In this way, the inventory of U-233 can be brought up from zero to the desired amount for regular operations. Bowman estimates a time period of six to twelve months for the particular system he describes to "boot-strap" itself up to full power. It should be noted that there would be a cost to undertaking this due to the electricity requirements of the accelerator.

> One of the greatest obstacles facing nuclear energy is how to properly handle the highly radioactive waste which is generated during irradiation in reactors. In order for nuclear power to realize its full potential as a major energy source for the entire world, there must be a safe and effective way to deal with this waste.[407]

The ATW subcommittee of the U.S. DOE Nuclear Energy Research Advisory Committee has explicitly stated that one of the four goals of ATW should be to "improve long-term prospects of nuclear power."[408] The two quotes at the beginning of this report clearly indicate the role that transmutation is to play in both revitalizing nuclear energy research and design efforts and in improving the viability of nuclear power in the future.

Even if the ultimate goal were phase out of nuclear power, implementation of separations and transmutation would result in a significantly increased nuclear infrastructure and guarantee the role of nuclear power for decades or even a century or two. It is obvious that any transmutation scheme would likely involve the expansion of MOX light water reactors, MOX or minor actinide fast reactors and possibly accelerator based systems (see above). New reprocessing facilities and reactors would have to be constructed and operated. Due to the need to recoup investments, and because of the waste penalty incurred if the program is halted partway through, it would be necessary to have full-scale implementation of transmutation. The timeframe estimated for various programs depends on the amount of material to be processed and the particular mix of technologies chosen. Research, design, development, and implementation are expected to take 118 years in the case of the U.S. ATW program. The Nuclear Energy Agency status report estimates a timeframe of 50-70 years to achieve an isotopic equilibrium for the plutonium and minor actinides.[409] This is for a mix of reactors corresponding to a capacity of 100 GWe.[410] According to the NEA, it would also be possible to progressively shut-down the reactors, starting with the light water reactors followed by MOX reactors and then followed by fast reactors fueled by actinides. In each stage, the residual actinides would be transferred to the remaining reactors. However, as the NEA notes, this would take "several decades or even centuries."[411]

Furthermore, it must be noted that many of the proposals for fast reactors are based upon reactors initially designed for breeding plutonium. Thus, these reactors could be switched from plutonium consumers to plutonium breeders at a future date.

Waste Management

Given the implications of instituting large-scale programs for waste transmutation, it is necessary to take a detailed look at the impact such programs would have on waste management. Spent nuclear fuel reprocessing and partial transmutation would significantly change the amount and nature of the waste being sent to a repository

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[407] IAEA 1997a, p. [3]
[408] Richter et al. 2000, p. 4
[409] OECD-NEA 1999b, p. 199. As fresh plutonium is put into reactors it is both fissioned and transmuted into minor actinides. After a period of time an equilibrium will be reached whereby the relative amounts of different isotopes of Pu and minor actinides will remain constant. This implies of course, a constant inventory of actinides in the reactors.
[410] OECD-NEA 1999b, p. 195. This is about equal to the installed nuclear capacity in the United States
[411] OECD-NEA 1999b, p. 204. Of course, there will remain residual actinides that must be placed in a repository.

and to disposal as low-level waste. The exact composition of the waste resulting from transmutation operations will depend upon the particular mix of separation and transmutation technologies chosen.

Reprocessing waste

As all transmutation proposals must rely upon extensive reprocessing operations, this will result in the generation of larger volumes of waste classified as low-level and intermediate level (TRU waste in the U.S.). Some of the increase in low-level waste production may be offset by decreases in low-level waste production in other steps in the fuel cycle due to decreased uranium processing requirements. However, the production of intermediate level waste from reprocessing (as well as fabrication of actinide fuel) is unique to these fuel cycles. A fuel cycle based on PUREX reprocessing and one pass of the plutonium through LWR reactors is estimated to approximately double (88%-115%) the volume of intermediate level waste to be disposed of in comparison to the once-through fuel cycle where intermediate level waste is only produced from reactor operations.[412] Multiple reprocessing of the plutonium fuel (again using PUREX and LWRs) results in an increase in intermediate level waste of 123%-149%.[413]

Thus, extensive reprocessing and actinide fuel fabrication associated with transmutation will result in increased volumes of process waste, particularly waste classified as intermediate level waste, which contains plutonium and other actinides. This will add to both the cost of transmutation and to the radiological risk of transmutation proposals.

Low Level Waste

As noted above, reprocessing and fuel fabrication activities result in the production of waste classified as low-level or intermediate-level. However, some transmutation proposals would also reclassify some of the radionuclides extracted from spent fuel as low-level waste. In particular, any transmutation proposals which seeks to either eliminate a repository altogether or dramatically increase its capacity by storing the medium-lived fission products until they decay, must rely on being able to dispose of significant portions of the waste as low-level waste.

There are two ways proposed in the transmutation literature for the residual waste (which includes long-lived fission products and actinides) to meet the regulatory limits of low-level waste. First, is to achieve high enough separations for the radionuclides of concern (e.g. plutonium, americium, technetium, and iodine) and then essentially close to 100% transmutation.[414] As this requires high transmutation rates for the actinides, this option requires the use of very advanced technologies based on accelerator systems. It also requires the above-ground storage of the medium-lived fission products for long periods of time. Setting aside all of the other issues that pose problems for transmutation (e.g. proliferation, safety, cost), these proposals ignore some of the basic practical difficulties with separating and transmuting the medium and long-lived radionuclides of

[412] Chow and Jones 1999, p. 37
[413] Chow and Jones 1999, p. 39
[414] See for example, Bowman 1997.

concern. These include the actual efficiencies achievable in both separation and transmutation for those radionuclides that can be transmuted and the long list of radionuclides that cannot be transmuted. There is no indication how all of the difficulties related to separation and transmutation of radionuclides such as Cs and Se (among others) would be overcome. Therefore, simply achieving high separations and transmutation efficiencies is insufficient and another method must also be used for the waste to meet low-level waste regulatory requirements.

The second method is to dilute the waste of the residual long-lived fission products and those radionuclides that cannot be transmuted so that they meet the regulatory limit.[415] Again, this requires highly efficient separation and transmutation of the actinides which dictates that an accelerator based system be used. It also requires, unless very large dilution volumes are used, the separation of the medium-lived fission products with above-ground storage for long periods of time. This is because these medium-lived fission products have the most stringent concentration requirements in the low level waste regulations and would therefore require the greatest amount of dilution. One justification that is made for this option is that stabilization of low-level waste is required by the U.S. regulations, which will result in dilution. Thus, the argument is that since LLW requires stabilization and stabilization is dilution, then dilution is permitted by the regulations.

The fact that either of these measures would be necessary in order for the waste to meet low-level waste regulatory limits clearly indicates that the waste, as initially processed, would not meet those limits. However, implementation of either of these proposals would be a violation of the low-level and other nuclear waste regulations and pose an undue threat to the environment and human health. First, it should be noted that if US regulations are to be used as the standard of comparison, the regulations are clear that waste from reprocessing of irradiated spent fuel is automatically considered high level waste, to be disposed of in a repository. According to the U.S. *Code of Federal Regulations* (10 CFR 60.2), high level waste is defined as:

> (1) Irradiated reactor fuel, (2) liquid wastes resulting from the operation of the first cycle solvent extraction system, *or equivalent*, and the concentrated wastes from subsequent extraction cycles, *or equivalent*, in a facility for reprocessing irradiated reactor fuel, and (3) solids into which such liquid wastes have been converted. (emphasis added).

In other words, the radionuclides extracted from spent fuel that are considered waste (i.e. not the plutonium or uranium) are automatically classified as high level waste.

Second, the possibility of storage above ground for up to 600 years currently has no basis in US regulations and the possibility of changing the regulations would have to be considered uncertain. As noted above, the *Code of Federal Regulations* clearly states that 100 years of institutional control should be considered the maximum feasible (10 CFR 61.59(b)) for waste disposal. In the case of storage of spent nuclear fuel (for example in a Monitored Retrievable Storage facility), the Nuclear Regulatory Commission regulations (10 CFR 70) allow for an initial license of only twenty years (renewable at the NRC's discretion). These regulations indicate that the general trend in management of nuclear waste in the United States is to assume a relatively short time

[415] See for example, Rubbia et al. 1997b.

frame between waste generation and eventual disposal. Even without such a regulatory stricture, the possibility of maintaining both the physical integrity of the waste and institutional control over the waste for time periods of over a century must be considered to have extremely high uncertainties and risks. Unfortunately, there is no clear answer to how long can be considered reasonable for long-term storage.

Third, dilution is not an acceptable waste management strategy, for any type of waste. According to the U.S. regulations, stabilization is meant to ensure that the waste has structural stability and will maintain its physical dimensions and form under anticipated disposal conditions (10 CFR 61.56(b)(1)). This can be achieved by the waste form itself, processing waste in order to create a more stable waste or by waste containers. A further requirement is that the void space in the waste be minimized if possible (10 CFR 61.56(b)(3)). This is a clear indication that the intent is to minimize waste volumes, not maximize them.

Thus, storage of fission products for hundreds of years and disposal of extracted fission products, and of residual radionuclides from transmutation, as low-level waste would appear to be in violation of at least three aspects of the Code of Federal Regulations governing disposal of radioactive wastes.

Just as importantly, however, the transference of waste to low-level waste disposal facilities is, in and of itself, problematic. This is because the regulation and disposal of waste classified as low-level is not based on a solid technical foundation to protect the environment and public health. This issue has been examined in detail by the Institute for Energy and Environmental Research in its book, *High Level Dollars, Low-Level Sense*.[416] However it is worth reviewing some of the findings of that study:

- The current regulatory definitions of radioactive waste are inappropriate. In particular, high-level waste and low-level waste are defined according to process that created them (with low-level waste actually acting as a catch-all for any waste not otherwise defined) rather than according to their actual radioactivity levels and the dangers they pose. This can result in situations in which waste classified as low-level is actually more radioactive than waste classified as high-level.[417] It can also result in situations in which long-lived radioactive wastes are disposed of as low-level waste.
- The regulations are internally inconsistent such that the limits set for low-level waste do not match the disposal requirements. For example, in some cases if waste that has been disposed of under institutional control for the maximum 100 years were to be dug up and re-buried it would require institutional control for another 100 years. In other words, it has not decayed to a level at which it no longer poses a concern as assumed in the regulations.[418]
- Low-level waste disposal sites have not performed as well as expected. At the time that the report was written, in 1992, three of six low-level waste disposal sites had been closed and had been expected to be maintenance free. Instead all three sites

[416] Makhijani and Saleska 1992.
[417] Makhijani and Saleska 1992, p. 117.
[418] Makhijani and Saleska 1992, p. 118

have required post-closure maintenance and clean-up activities due to radionuclide migration within ten years of closing.[419]

These illustrate some of the problems with the current regulation of radioactive wastes and, in particular, the problem with waste classified as low-level waste. These issues must be accounted for in proposals for transmutation. In other countries, the regulation of radioactive waste may not face the same problems and may be based on a more technically justifiable foundation. However, in those cases it seems unlikely that transmutation waste could be reclassified as low-level waste. For example, Swedish radioactive waste management is based upon the longevity of the waste. As a result, Swedish waste management plans call for disposing of reactor waste which would be considered low-level in the United States, in a geologic repository (this accounts for 40% of the volume of waste to be sent to a future repository in Sweden).[420] Thus, it is unlikely that such a system would allow radionuclides such as Cs-135 or Se-79 to be disposed of as low-level waste. In France and Britain, nuclear waste classifications are also based upon longevity and the characteristics of the waste rather than its origin.

Temporary storage of medium-lived fission products for long time periods in order to allow them to decay to the limits set for low-level waste, as well as the disposal of long-lived fission products as low-level waste, should not be considered an acceptable form of waste management. Disposal of medium and long-lived fission products that cannot be transmuted as low-level waste transfers the same amount of radioactivity from a deep geological repository to shallow land burial. While potentially reducing repository doses it will result in increased doses from the LLW waste burial. No comparison has been made of the overall risk of transferring these materials to LLW disposal. Given the significantly reduced requirements for isolation of radioactivity from the environment, even for Class C low level waste (the highest classification of low-level waste with the most stringent requirements), the risks from such a practice are of serious concern.

Timeframe for Implementation

Implementation of transmutation programs will take a significant period of time. Even the simplest of proposals to extend MOX usage and add fast reactors for higher levels of actinide fissioning are unlikely to come into fruition for another two decades.[421] Proposals for full scale transmutation (either based entirely on accelerators or based on both reactor and accelerator systems, often called dual-strata systems) have even longer time horizons. This raises a number of serious questions, as programs for solidifying liquid high level waste and repository siting, construction, and emplacement will all be ongoing:

[419] Makhijani and Saleska 1992, p. 69
[420] Makhijani and Saleska 1992, p. 119
[421] OECD-NEA 1999b, pp. 182-183. In one scenario, transmutation of Np and Am in PWRs would start in 2010, but on a modest scale. Another would use PWRs until 2020 when fast reactors would start contributing.

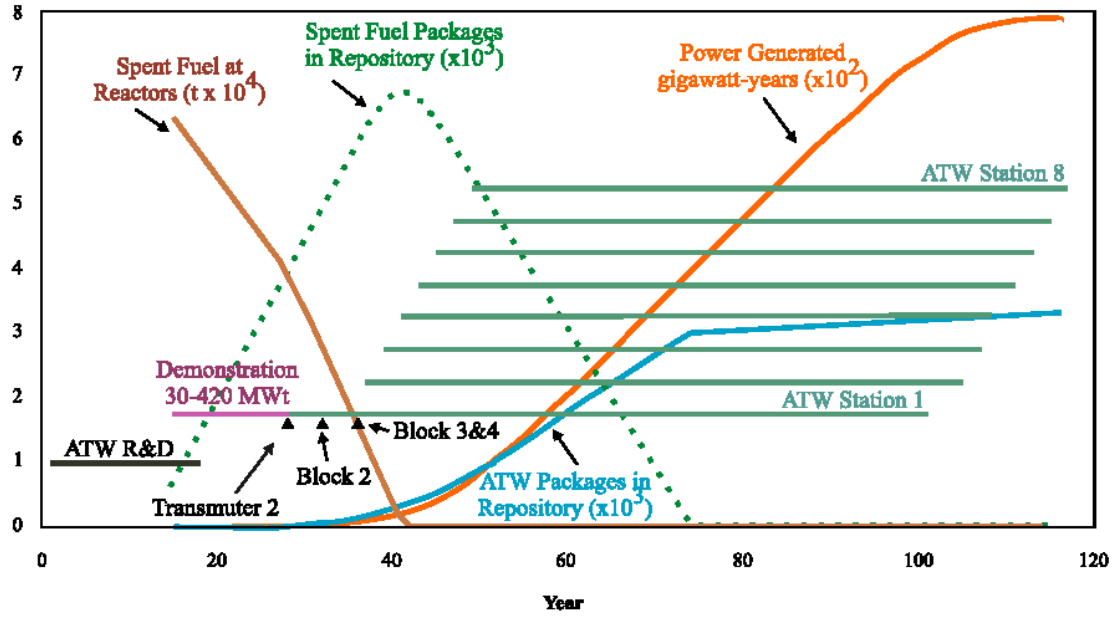
- Will waste already emplaced in a repository be removed for processing for transmutation?
- Will reprocessing waste which has been immobilized be processed again?

The ATW Roadmap provides some interesting information in the case of U.S. implementation of transmutation. The Yucca Mountain repository is scheduled to begin accepting waste in 2010. The emplacement of spent fuel and other high level waste in Yucca Mountain would end around 2035, the same time that full-scale transmutation would begin to occur assuming an immediate start to the R&D program. The Roadmap report states "Thus, repository development and waste emplacement activities could be completed before the first ATW station would start operations. Because ATW stations will require a repository for disposal of ATW waste forms, from a waste management perspective it is appropriate for the current repository development program to proceed as planned."[422] In other words, if Yucca Mountain is opened as the US high level waste repository, at lease some of the spent fuel will be shipped from reactor sites all over the country to Nevada. Then, it will be taken out again and shipped to one of eight transmutation stations. Finally, the transmutation waste will be shipped back to Nevada for placement back into the repository. This can be seen in Figure 22 which shows the amount of spent fuel in the repository rising to a maximum and then declining as it is removed for transmutation. At the same time, the amount of ATW high level waste being shipped back to the repository begins to rise.

As noted in the ATW Roadmap report, if waste already emplaced in the repository has to be removed for shipment off-site (and then back to the site for re-emplacement after transmutation), there would be an increased risk to both repository and transportation workers, as well as to the public.[423]

[422] ATW Roadmap 1999a, p. 5-6
[423] ATW Roadmap 1999a, p. 5-6

Figure 22: Integrated Schedule for RD&D and Deployment of ATW Technology

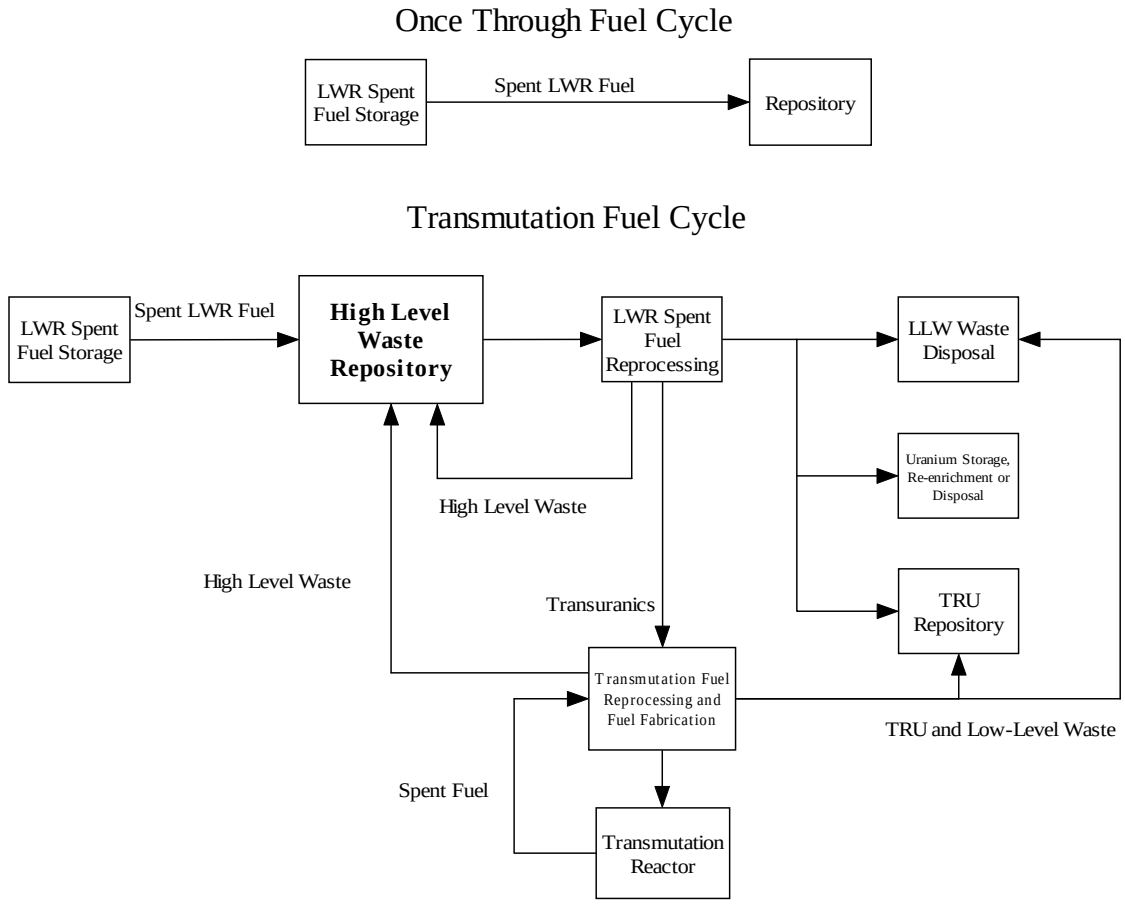


Source: Figure 7.1 of ATW Roadmap 1999a.

In general, any transmutation proposal would require increased transportation of spent fuel and radioactive waste. A flow-chart of this process of increased transportation can be seen in Figure 23.[424] This is a generic diagram and does not correspond to any particular set of transmutation technologies or any particular transmutation proposal. As a result, some of the steps seen here might not be necessary. For example, if spent light water reactor fuel is sent directly for reprocessing rather than sent to the repository first, that would eliminate that intermediate step. Also, this diagram shows separate facilities for processing of light water reactor fuel, transmutation reactor fuel fabrication and reprocessing, and transmutation reactors. This would be the case if a centralized facility is used for reprocessing LWR fuel and a centralized facility is used for reprocessing fuel from the transmutation reactors (so that one reprocessing facility would service multiple transmutation reactors). Another alternative would be for all of these facilities (or the transmutation reprocessing and transmutation reactors) to be collocated on the same site. This would avoid transportation between separate sites (however, with an economic and possibly other penalties as a result of having multiple smaller reprocessing facilities).

[424] Adapted from Figure 6-1 of NAS-NRC 1996, p. 103

Figure 23: Transportation For Once-Through Versus Transmutation Fuel Cycles



Geologic Repositories

The claim has been made about transmutation that it could eliminate the need for a repository entirely.[425] This is based upon the idea that with efficient enough separations and transmutation of the actinides, technetium, and iodine, and storage of hundreds of years to allow the medium-lived fission products to decay, the residual waste would meet Class C requirements for disposal as low-level waste. In fact, one proponent of transmutation even stated that the waste could be fabricated into concrete blocks, buried five feet underground (as required by Class C) and then a parking lot for the plant built above.[426] This of course seems to ignore the fact that some of the materials would have to be stored for several hundred years before meeting the Class C requirement (even if the assumption is accepted that a repository is not required).

While some may continue to see elimination of a repository as the ultimate goal of transmutation, it has come to be generally recognized that this will not be achieved. This has been recognized by the U.S. National Research Council, the International

[425] See for example, Bowman 1997 and Rubbia et al. 1997b, and NAS-NRC 1996, p. 243 and p. 287. The National Research Council references are for proposals by researchers at Los Alamos National Laboratory (LANL) and Brookhaven National Laboratory.
[426] Bowman 1997a, pp. 140-145

Atomic Energy Agency, and the Nuclear Energy Agency of the Organization for Economic Cooperation and Development (NEA-OECD).[427] At best, transmutation may change the amount and nature of the waste to be placed in a repository. However, to have any significant impact on the loading of the repository, both the volume of the waste and the radioactivity of the waste would have to be significantly reduced. However, even this may not be achievable. The effect of transmutation on repository programs will be discussed in the context of three parameters: the capacity of the repository, the performance of the repository, and the dose from the repository. It must be recognized, of course, that there are a number of repository programs underway worldwide. It must also be recognized that while some transmutation proposals are made in the context of a gradual phase-out of nuclear power operations, most assume a continuation of nuclear power. Thus, nuclear waste would be continually produced with no endpoint in sight. Therefore, this section will attempt to provide generic information about the effect of transmutation on repository programs with specific examples where possible.

Implementation of transmutation proposals could also exacerbate many of the long-standing institutional and political issues associated with the management of nuclear waste.[428] However, for the purposes of this report, we will examine the management of nuclear waste from transmutation under the prevailing assumption that a geologic repository is the waste management method to be used.

Repository Capacity

Unlike waste that is considered low-level or transuranic, the capacity of geological repositories for spent fuel and high level waste is set not by the volume of waste to be managed, but by its heat content. The decay of radionuclides in the waste generates a large amount of heat. A particular repository design will only allow a certain amount of heat within a unit of area.

Transmutation will have an effect on the capacity of any given repository. However, that effect could be to increase or decrease the capacity of the repository depending on the isotopic composition and heat generation of the final waste in comparison to the waste that would originally have been placed in that repository (e.g. spent thermal reactor fuel). It will also depend on the timeframe in which the waste is emplaced in the repository (longer timeframes result in lower heat, mainly due to decay of short and medium-lived fission products).

The ability of transmutation proposals to increase repository capacity is severely limited because two of the main contributors to decay heat, Sr-90 and Cs-137, cannot be transmuted. These medium-lived fission products (both the inventories contained in the original spent fuel or high level waste, plus that produced during transmutation from fissioning of the actinides) must be sent to the repository (as discussed above, the option of long-term storage is not viable). In addition, transmutation can result in the increased production of higher actinides that result in greater decay heat in comparison with plutonium. This is particularly the case with thermal reactors. For example, Chow and

[427] NAS-NRC 1996 p. 7, OECD-NEA 1999b p. 57, IAEA 1999 p. 6

[428] See Makhijani 1999 for more information on the current problems in radioactive waste management and possible solutions.

Jones from RAND find that the heat from the use of MOX (with one pass of the plutonium) has a higher heat content than the once-through fuel for nearly 1000 years due to the increase in Am-241 and Cm-244 in the fuel.[429] When plutonium is repeatedly reprocessed and passed through a thermal reactor, the results are a little more complicated, with the reprocessed fuel have an even higher heat content initially, but reducing to the level of spent LWR fuel sooner (in less than 100 years).[430] In that situation, the timing of emplacement becomes important.

The potential impact of more advanced transmutation systems on heat content of the waste being sent to a repository will depend greatly on the mix of technologies chosen (e.g. the particular mix of thermal critical reactors, fast critical reactors, and accelerator based systems) as well as the mix of radionuclides considered for transmutation. For example, if curium is not processed and transmuted in critical reactor systems, this will have a significant impact on the decay heat due to Cm-244, which will be produced in greater amounts. The fuel from fast reactors will be of particular concern due to the high actinide content (fast reactors are fueled at a higher percentage of fissile isotopes) and the long burn-up times which results in greater production of higher actinides. By one estimate, spent fast reactor fuels emits nearly twenty times more heat per ton of heavy metal as conventional uranium oxide fuel.[431] In fact, the fuel is so hot that if it is not put in a large repository with previously cooled LWR waste (essentially "diluted" according to the literature), it must be stored for hundreds of years before disposal or transmuted in an accelerator based system.

Again, however, it should be noted that even in systems that have a combination of thermal and fast reactors or in systems that are accelerator based, the original medium-lived fission products must still be accounted for in determining repository capacity. Thus, in all of these scenarios there is a definite possibility that repository capacity may actually decrease as a result of transmutation.

While the capacity of the repository is set by the heat content, it is also useful to consider the mass of waste to be handled. First, in some countries, such as the United States, there is a statutory limit on the amount of waste that can be emplaced (for the U.S. the first repository is limited to 70,000 metric tons of heavy metal).[432] Second, this is an indication of the scale of effort necessary to handle the waste streams.

One of the primary means that the mass of waste to be sent to the repository will be reduced is by separation of the uranium from the rest of the fuel. However, the fate of that uranium remains an open question. For example, under the proposed US plans for ATW, processing the 63,000 metric tons of commercial spent fuel currently slated to be sent to Yucca Mountain will result in 46,000 metric tons of high level waste (about 36,000 metric tons will be from processing the original spent fuel and 10,000 metric tons

[429] Chow and Jones 1999, p. 22.
[430] Chow and Jones 1999, pp. 24-29.
[431] OECD-NEA 1999b, pp. 214-215. Note that this is based on specific scenario and should not be considered an authoritative number. It should also be noted that the initial radiotoxicity of this spent fuel is 74 times that of ordinary light water reactor fuel.
[432] The 70,000 metric ton limit was set in the Nuclear Waste Policy Act of 1982, as amended (in 1987). The limit is not necessarily a technical limitation, but rather a legislative one. This legislative limit is in effect until a second repository is opened. DOE 1999b, p. S-8,S-9

will be from processing the ATW fuel).[433] Thus approximately 73% of the weight of the original spent fuel will still require geologic disposal after transmutation. However, the uranium that originally formed the bulk of the spent fuel must also be disposed of in an environmentally sound manner. The uranium has a mass of 59,600 metric tons. According to the ATW Roadmap, this uranium would be disposed of as Class C low-level waste.[434] This assumes that the uranium will meet the Class C limit. As was shown above, there is no justification for treating uranium as a Class C waste. Protection of the environment and human health dictate that the uranium be treated in the same manner as waste containing transuranic radionuclides. If this put into effect, the mass of waste requiring geologic disposal would rise dramatically from 63,000 metric tons to 106,000 metric tons.[435] Thus, instead of increasing the capacity of a repository, transmutation could in fact result in nearly 70% more waste requiring repository disposal.

The above comparison, however, does not take into account the fact that the ATW stations would be producing electricity at the same time as they transmuted some of the LWR spent fuel. If one assumes that LWRs would have otherwise produced all of that electricity then this avoided LWR spent fuel production must be essentially credited to the ATW system. Assuming standard parameters for current LWRs, this corresponds to approximately 18,000 MTHM of spent fuel.[436] In other words, in addition to the 63,000 MTHM of LWR spent fuel the ATW system would process, it would also avoid 18,000 MTHM of LWR spent fuel generation. However, even with this credit, the gains made by ATW are illusory. The high level waste generated by ATW would still be almost 60% of the original and avoided mass of spent fuel. Adding in the uranium would mean that ATW would increase the mass of material requiring repository disposal by about 30%.

Repository Performance

Reprocessing and transmutation would have two other effects on a repository. One is related to the increased dose from the repository due to higher loading (more waste can be placed in the repository) and is discussed below in the section on Repository Doses. The second is related to the performance of the repository due to the change in

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[433] For every 170 MTHM of spent fuel processed (the amount processed per year per station), a total of ~60 MT of ceramic waste (~24 m^3 per year) and ~65 MT of metal waste (~8 m^3 per year) is produced. This includes the waste produced from processing the resulting ATW fuel, which is also then processed (approximately 26 MT). ATW Roadmap 1999d, p. 28.

[434] We have used a nominal 94.6% to represent the uranium content in the spent fuel in order to be consistent with the ATW roadmap (ATW Roadmap 1999d, p. 37)

[435] This comparison is imprecise as it compares spent fuel in MTHM (which is only a measure of the weight of the heavy metals, as opposed to the spent fuel itself which includes, among other things, the cladding) and the pyroprocessing waste which is measured in metric tons (i.e. it is not only the weight of the heavy metals). However, this comparison will suffice for illustrative purposes.

[436] Each sub-critical reactor produces 840 MWt and there would be 8.5 stations, each with 8 reactors for a total of 57120 MWt. Over the course of sixty years of operation for each station and assuming an 80% capacity factor (which is high and therefore conservative), the total output would be 1.0×10^9 MWd. Assuming the LWRs operated at a burn-up of 40,000 MWd/MTHM, the total amount of spent fuel discharged from LWRs producing an equivalent amount of power would be approximately 25,000 MTHM. However, this calculation is for ATWs processing 87,000 metric tons of spent fuel rather than the 63,000 metric tons of spent fuel being discussed in the text. Applying a scaling factor of 63/87 results in a total avoided spent LWR fuel output of 18,000 MTHM.

the short term thermal profile of the repository from the possible removal of medium-lived fission products and actinides. For spent fuel, the fission products (particularly Sr-90 and Cs-137) dominate the thermal power for about seventy years when the thermal power from the fission products equals the thermal power from the actinides. Subsequently, it is the actinides that dominate the thermal power.[437]

Two distinguishing features of repositories that play a factor in the transmutation debate are the level of water and the temperature of the repository:

1. Saturated vs. Unsaturated: A saturated repository is below the groundwater table. Though there is significant water presence in a saturated repository, the water movement could be slow. An unsaturated repository is above the water table though rainwater seepage can result in significant water content within the pores of the rock.[438]

2. Hot vs. Cold: A hot repository relies on the decay heat of the waste to maintain temperatures above the boiling point of water. The purpose is to prevent corrosion of the waste canisters. In a cold repository, the temperature is below the boiling point. This is done by having a lower density of fuel emplacement, often accompanied by planned decades-long storage before emplacement to allow further decay of fission products. The thermal loading strategy of the repository is only concerned with the short-term temperature profile of the repository since the long-term temperature is governed by the long-lived radionuclides. The choice of thermal loading strategy is complex and involves consideration of corrosion of the canister, thermal stress of the rock which can make the repository less predictable, and ability of radionuclides to migrate through the rock.[439]

The thermal loading of the repository plays a significant role in the performance of the repository. Removing the majority of the actinides and/or the medium lived fission products significantly changes the heat output of the waste. Removal of all actinides (which is not expected under even the most optimistic projections) results in about a twenty percent drop in thermal power (at the ten year mark). Removal of both actinides and Sr-90, and Cs-137 results in a thermal power at ten years which is one percent of spent fuel.[440] On the other hand, if a combination of thermal and fast critical reactors is used, as is proposed in some limited transmutation schemes, the decay heat may actually increase. As discussed above, fast reactor fuel will be irradiated to much higher burn-up levels (i.e. it will stay in the reactor for a longer period of time) and have a higher actinide content to begin with. As a result, fast reactor spent fuel will emit heat at levels many times that of ordinary light water reactor spent fuel.[441] This could have a significant effect on the repository due to thermal stress on the host rock. It would either have to be

[437] NAS-NRC 1996, pp. 323-324

[438] Bodansky 1996, p. 145. Bodansky gives the example of one study which showed "that most of the pores in the rock in the unsaturated zone [at Yucca Mountain] are more than half filled with water, although fractures in the rock are usually drained by capillary action."

[439] Bodansky 1996, pp. 148-149.

[440] NAS-NRC 1996, p. 323

[441] See, for example, OECD-NEA 1999b, p. 214-215. The increase in heat emission will depend greatly on the particular isotopic mix of the various elements. In the example cited here, the authors found that the fast reactor spent fuel was 20 times hotter than a comparable amount of light water reactor spent fuel.

"diluted" by placing it within a much larger quantity of cooler light water reactor fuel or high-level waste, be stored in engineered facilities above ground for a period of hundreds of years, or further processed in accelerator based systems.[442]

The effect of this change in thermal power on the repository performance will depend on the exact geological conditions of the repository site and much of the information necessary to make detailed determinations is not available.[443] However, the National Research Council has come to some generic conclusions related to the decrease in heat from removal of actinides and medium-lived fission products, based upon the general characteristics of saturated and unsaturated repositories.

In a saturated repository, one in which the repository is flooded sometime after closure, the lack of heat could actually have some positive effects on limiting the transport of radioactive materials. This is because high temperatures, under certain conditions, can have unwanted effects on materials, increase radionuclide solubilities, and decrease radionuclide migration retardation by natural ion exchange materials.[444] The stresses on the surrounding rock can also create pathways for water flow and temperature differences can act to drive water flow. However, the precise effects and their impact on repository performance are currently unknown. Thus, the NRC concludes that reduction of the thermal power should have an overall beneficial effect.[445]

On the other hand, the effects on an unsaturated repository of reducing the heat load could be quite harmful. In an unsaturated repository it is necessary to keep the waste containers dry to avoid corrosion due to moisture. One method to accomplish this is to use the decay heat of the spent fuel. By determining the optimal thermal pulse, the waste containers can theoretically be kept dry for an extended period of time (over 10,000 years according to some). This prevents water contact with the waste container. This is currently the preferred option for Yucca Mountain.[446]

There remains a significant amount of work to be done to determine optimal heat loading for repositories and much of the relevant information is not yet available. However, if it is determined that high heat loading for a particular repository is desirable, this may not be possible after transmutation.

Repository Doses

One of the main stated arguments for implementing any transmutation program is to reduce the dose from the geologic storage of high-level waste. Specifically, the goal is to reduce inventories of those radionuclides that pose the largest dose risks. This has

[442] OECD-NEA 1999b, p. 214-215.
[443] NAS-NRC 1996, p. 326
[444] Certain materials when exposed to a solution containing ions (i.e. positively or negatively charged atoms) will absorb those atoms and release an ion present in the original material. Thus, the migration of certain radionuclides could be slowed down because such ion-exchange materials absorb them.
[445] NAS-NRC 1996, p. 327. There was one general exception noted. Under certain conditions, the solubility of certain actinides may actually decrease under high heat conditions. If these particular actinides dominate the repository risk, then a reduction in the thermal power could actually increase the risk from the repository. However, the NRC concluded that it was unlikely that conditions would exist such that actinides that exhibited "retrograde solubility" were found to dominate the repository risk.
[446] NAS-NRC 1996, p. 328

resulted in a concentration on the actinides (due to their high radiotoxicity) and those long-lived fission products expected to dominate the repository dose.[447] For example, in the United States, the focus is on technetium, iodine, and neptunium, which dominate the Yucca mountain repository dose over the long-term. However, as noted in Chapter I, the solubility, and hence the health impact of technetium and neptunium are directly related to the choice of repository.

The effect of transmutation on the expected dose from any repository program is highly variable. It will depend on such factors as the scale of the transmutation program (e.g. MOX vs. MOX plus fast reactors vs. ATW) and the key radionuclides of concern for the particular repository.

The discussion of the dose benefits of transmutation often focus on the reduction in the amount of plutonium and minor actinides in the waste since these isotopes have the largest radiotoxicity of any group in spent fuel. However, in terms of assessing the long-term doses to the public from a repository it is as necessary, if not more necessary, to consider the long-lived fission products and activation products. This is due to the fact that the dose will be determined not only by the inventory of the radionuclides, but also their transport in the environment. The precise nature of the transport of radionuclides in the environment continues to be an area of investigation. It is generally stated that certain radionuclides among the long-lived fission products travel quite quickly through the biosphere while some of the actinides may be strongly retarded by the geological media. However, environmental monitoring data has shown that plutonium can travel quite rapidly (for example at the Nevada Test Site near Yucca Mountain) and recent plutonium chemistry experimental results indicate that plutonium oxides may be significantly more soluble than previously assumed.[448] Radionuclides are therefore sometimes given one of three classifications:

- Inventory Limited: The transport of these radionuclides (such as iodine-129) in the environment is primarily limited by the amount of the radionuclide present in the waste. Reducing the amount of the radionuclide consequently reduces the dose from the radionuclide.
- Solubility Limited: The transport of these radionuclides (usually the actinides) in the environment is limited by the rate at which they dissolve in water and whether they bind to the geologic media at the repository site. Reducing the inventory of these radionuclides is not expected to have a large impact upon the dose.
- Variable: These radionuclides (including important radionuclides such as technetium-99 and neptunium-237) vary greatly in their transport through the environment depending on the specific conditions of a repository. Thus, Tc-99 can be inventory limited in an oxidizing repository such as Yucca Mountain, but may be solubility limited in a repository with reducing conditions.

[447] As noted elsewhere, most repository models assume that the actinides will have their migration significantly retarded and they will thus contribute less to the overall repository dose despite their higher radiotoxicity in comparison to the long-lived fission products.
[448] See Kersting et al. 1999, Haschke, Allen, and Morales 2000and Madic 2000

An analysis of the key radionuclides for spent fuel, high level waste and high level waste from fast reactors has indicated that the dose for ground-water release from a hard rock repository is dominated by ^{126}Sn, ^{226}Ra, ^{129}I, ^{230}Th, ^{135}Cs, ^{79}Se, ^{14}C, ^{36}Cl, and ^{233}U. The estimates for salt and clay repositories are similar.[449] A comparison of relative estimated doses from unreprocessed spent fuel in a granite repository in Sweden, Finland, and the United Kingdom shows similar results with the addition of ^{99}Tc as another important radioisotope.[450] For all the different repositories, neptunium-237 and americium-241 may be considered important, both in terms of their direct effect on dose (for example, Np-237 is important at Yucca Mountain) and for their production of U-233 through radioactive decay.[451]

The effect of repository choice is particularly important in determining the applicability and efficacy of any transmutation program. As noted in the MIT Review of ATW:

> Technetium and neptunium are targeted due to their solubility in the oxidizing conditions at Yucca Mountain. The problems associated with these elements stem from the repository choice, not their inherent chemical behavior. Selection of a repository with reducing conditions would eliminate the need to address these elements since they would be insoluble in the tetravalent state. Care should be taken in future presentations to explain that the concern is due to oxidizing conditions at Yucca Mountain.[452]

In other words, the justification for transmuting technetium and Np-237 in the US program is primarily dependent upon a decision to build a repository at Yucca Mountain. The Yucca Mountain repository faces numerous problems, including its oxidizing conditions, and has been criticized as a poor choice.[453]

Examining the list of key radionuclides and comparing it to the list of radionuclides for which transmutation is possible clearly indicates the limitations of transmutation. Only Np-237, Am-241, Tc-99, and I-129 are included in the list of radionuclides to be transmuted, and then only in certain programs. Uranium and radium would be indirectly reduced due to the inclusion of their parent radionuclides. However, the rest will be sent to the repository with no change in inventory, released during reprocessing operations or separated from reprocessing waste and disposed of as low-level waste.

At the same time that some radionuclides are being transmuted, there is also the problem of a shift of the minor actinides to higher numbers due to neutron absorption. This may be particularly an issue in the case of transmutation programs relying solely on a combination of MOX-fueled light water reactors and actinide fueled fast burner reactors. Such programs, relying on critical reactors, are limited in their ability to achieve high transmutation rates. As a result, while inventories may be reduced, the

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[449] Volckaert et al. 1999, p. 465-467
[450] NAS-NRC 1996, p. 330
[451] Volckaert et al. 1999, p. 466
[452] Kazimi et al. 1998.
[453] A review of the Yucca Mountain repository is beyond the scope of this report. For more information concerning IEER's critique of the Yucca Mountain project, readers are invited to visit the IEER website (http://www.ieer.org) where they can find numerous sources, including articles in *Science for Democratic Action*, comments submitted to various government agencies, and a 1992 report on radioactive waste management (Makhijani and Saleska 1992).

remaining waste may be more radioactive. For example, a comparison of LWR spent fuel, LWR high level waste, MOX high level waste and fast burner reactor high level waste reveals that the peak dose from fast burner reactor HLW is larger than that for either LWR or MOX fuel or HLW.[454] In part this may also be due to the fact that the actinides are generally limited in their impact by their solubility and transport, not by their inventories. Thus, reducing the inventory of plutonium and other actinides may not actually have much of an effect on reducing repository doses.[455] Reduction in the inventory of actinides would have an effect, however, on the dose from inadvertent intrusion into the repository.

It has also been suggested that transmutation may actually increase the long-term individual dose from a first repository in those countries requiring more than one repository to handle the waste.[456] One of the main advantages cited by transmutation advocates is the increase in the amount of waste that can be placed in a repository if the high-heat medium lived fission products are removed. While this may be true if the medium lived fission products are stored outside the repository, it also means that any repository will then end up with a higher concentration of those elements that must be placed in a repository, namely the residual actinides and long-lived fission products. As a result, the source term for these radionuclides will increase and thus the doses from that repository will be higher.[457] For example, Table G-5 of the National Research Council's report on transmutation shows that the peak dose rate from I-129 and Tc-99 in an unsaturated tuff repository would increase with separation of Sr and Cs. Again, this is because a greater amount of waste could be placed in the repository. This table also shows that the doses from Np-237 and Pu-242 would also increase with removal of the medium lived fission products, despite the fact that the inventory of these radionuclides would be lower. This is because a larger number of waste containers could be placed in the repository (since each one would not be as hot). This results in a larger number of waste containers that can fail and a higher surface area of waste containers in order for the radionuclides to come into contact with water. Because the release of neptunium and plutonium is limited by their solubility, the increase chance of contact with water is a major factor.[458]

Another factor affecting doses from a repository is the radionuclide inventory of individual containers of spent fuel or high level waste. The degree to which radionuclides enter the environment is based, in part, on the failure of individual waste

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[454] Volckaert et al. 1999, p. 466

[455] NAS-NRC 1996, p. 342.

[456] NAS-NRC 1996, p. 343. For example, in the United States, the first repository is limited to 63,000 metric tons of commercial spent fuel and high level waste. However, as has been noted elsewhere in this report, a total of 87,000 metric tons of waste is expected to be produced under current reactor licenses. Thus, either the restriction on the first repository must be lifted or a second repository will be necessary. These figures do not include defense spent fuel and high level waste, which must also be disposed of in a repository.

[457] NAS-NRC 1996, p. 343. Of course, the purpose of higher waste emplacement is to avoid a second repository, which would have its own associated dose. However, with removal of medium lived fission products, it would mean that the source term would all be in one repository instead of two.

[458] NAS-NRC 1996, pp. 343-344. The NRC panel noted that more conservative solubility assumptions for neptunium-237 would result in a much higher dose from this radionuclide (perhaps by a factor of 100,000 or more).

containers. For example, the inventory of higher actinides in high-burnup fast reactor fuel is significantly higher than in LWR fuel.[459] Thus the failure of a container of fast reactor spent fuel may have a greater effect than the failure of an LWR spent fuel container. This must be considered in determining how doses from a repository will be affected by transmutation. In other words, not only might there be more waste overall in the repository and more waste containers that can fail, the consequences of the failure of any individual waste container might be higher than before.

Of course, if transmutation means avoiding construction of a second repository, then there would not be a dose to the population near the second repository site. In other words, the collective dose (for those populations at both repositories) may go down, but the individual dose to a person at the first repository may increase. The equity of avoiding doses in one location (the second repository) by increasing doses at another (the first repository) would compound the already inherently unjust burden placed upon the population near the repository.

The ATW Roadmap provides a good estimation of the best case scenario for the effect of transmutation on repository doses. It assumes near perfect elimination of the actinides and long-lived iodine and technetium from the commercial and defense spent nuclear fuel. Also, the waste is assumed to be disposed of in Yucca Mountain where technetium, iodine, and neptunium dominate the dose. However, the defense high-level waste (both spent fuel and liquid waste) is assumed not to be processed because of the difficulty in processing this waste. Using the models for Yucca Mountain that have previously been developed, the ATW Roadmap compares the dose from disposal of ATW high level waste to the reference case of direct disposal of spent nuclear fuel in Yucca Mountain. Under this scenario, the dose from ATW is about four orders of magnitude (10,000 times) less than the reference case until about 8,000 years.[460] However, the difference is dramatically reduced after 8,000 years due to the contribution of the defense waste (both spent fuel and high level waste). At the time of the peak dose, approximately 300,000 years, the ATW case is only about ten times lower than the reference case. Even with the addition of Defense spent nuclear fuel transmutation, the final reduction in dose is calculated to be two orders of magnitude (100 times) at the time of peak doses.[461] An assessment of reducing the total number of waste packages was also conducted. The number of waste packages was reduced by a factor of ten. The peak dose does not change from the ATW scenario with a larger number of packages. However, in the time period of about 3,000 to 12,000 years the dose increases in comparison with the standard ATW scenario (by as much as a factor of ten).[462]

Table XX of Chapter I provides an overview of the amount of spent fuel, vitrified high level waste, and liquid high level waste for two countries (the U.S. and France). If separation of high-level liquid and vitrified waste proves to be significantly more difficult or expensive that separation of spent fuel, then it is more likely that only spent fuel will be processed for transmutation. As can be seen from Table XX, this could have a

[459] OECD-NEA 1999b, p. 215.
[460] ATW Roadmap 1999c, p. 3-1
[461] ATW Roadmap 1999c, p. 4-1
[462] ATW Roadmap 1999c, pp. 3-3 – 3-4.

significant impact on the dose reduction of transmutation, particularly for those countries with significant stocks of reprocessing waste.

The case of Yucca Mountain provides the perfect example for the need to change nuclear waste management policies. The selection of Yucca Mountain by Congress before the Energy Department completed all of the preliminary technical work has resulted in the spending of billions of dollars on an unsuitable repository. At first glance, transmutation would seem to be well suited to reducing the risks from the Yucca Mountain repository. The three radionuclides with the largest impact on the dose due to the geology at Yucca Mountain (Tc-99, I-129, and Np-237) are considered candidates for transmutation. The presence of resources (including precious water resources in an area where water is scarce) would seem to indicate that transmutation could reduce the potential impact of human intrusion. What is overlooked in that argument, however, is that it also makes a strong case for Yucca Mountain to be abandoned from active consideration as a repository. A site without Yucca Mountain's geological conditions favoring solubility and transport of key radionuclides, without the need for high heat loading to prevent corrosion and without regionally rare and precious resources (such as water or certain minerals) could also result in a significantly lower repository dose without all of the disadvantages of separations and transmutation. In order to adequately assess the potential of transmutation, this reduction in dose must be compared to the possible reductions in dose that would result from other changes in waste management practices (for example, if the repository were sited at a different location). As far as can be determined from the available literature, such a comparison has not been made.

While it is useful to estimate the possible reductions in dose due to transmutation, it must be done in the context of an overall risk reduction approach. Such an approach must look at the following factors in an integrated fashion:

- Repository Dose: Repository doses may decrease or they may increase under different transmutation programs. This will depend on which radionuclides are transmuted, to what extent they are transmuted and the conditions of the particular repository.
- Reprocessing Dose: In addition to the potential increase in worker doses due to reprocessing, there may some more long-term and/or wide-spread effects of reprocessing. Some of the key radionuclides such as I-129, C-14 and Cl-36 are ignored in repository dose comparisons because they are either released to the environment during reprocessing or end up in other waste streams (e.g. low-level or intermediate level waste).
- Activation Products: Neutron activated materials, as well as spallation products in the case of accelerator based systems, would have to be assessed for their potential health effects. The current practice of disposing of decommissioned reactors in shallow-land burial may not be appropriate in all cases. Components subjected to the high neutron flux of ATW in particular need to be properly accounted for in any calculations.
- Extracted Uranium: Doses from extracted uranium, if it is disposed of as low-level waste, could be high and comparable to repository doses. As is discussed above, extracted uranium should be treated in the same manner as TRU waste. However, the

> uranium also poses a risk from build-up of both Pa-231 and Ra-226. Transmutation of their transuranic parents would reduce their long-term production. However, both radionuclides would still be produced as the result of the decay of U-234 and U-235. If the uranium is disposed in shallow land burial, the doses from these two key radionuclides could be greater than if they were disposed of as part of spent fuel.[463]

A comprehensive evaluation of the radiological risks of transmutation would account for at least these factors. Otherwise, reductions in repository doses (assuming that is the result) would be done at the expense of increasing doses from other sources. Such a shift in risk, and potentially an increase in risk, would be hard to justify.

Given that transmutation may produce very mixed results in terms of reducing the dose from the repository and may increase the risk from other sources it is necessary to ask if there is another means to achieve comparable results. Elsewhere IEER has recommended a program for long-term radioactive waste management that would entail storing spent fuel and research on geologic repositories and other alternatives

[463] NAS-NRC 1996, p. 333-334

Appendix A: Basics of Nuclear Physics

Reprinted from *Nuclear Power Deception: U.S. Nuclear Mythology from Electricity "Too Cheap to Meter" to "Inherently Safe" Reactors*, by Arjun Makhijani and Scott Saleska, Apex Press, 1999, pp. 207-214

Appendix A: Basics of Nuclear Physics

Structure of the Atom

The atoms of which every element of matter is composed have a nucleus at the center and electrons whirling about this nucleus that can be visualized as planets circling around a sun, though it is impossible to locate them precisely within the atom. The nuclei of atoms are composed of protons, which have a positive electrical charge, and neutrons, which are electrically neutral. Electrons are electrically negative and have a charge equal in magnitude to that of a proton.

The number of electrons in an atom is normally equal to the number of protons in the nucleus. As a result, atoms of elements are normally electrically neutral. The mass of an atom lies almost entirely in its nucleus since protons and neutrons are far heavier than electrons.

Free neutrons are unstable particles which decay naturally into a proton and electron, with a half-life of about 12 minutes.

neutron ===> proton + electron + a neutrino

However, it is remarkable that neutrons, when they exist together with protons in the nucleus of atoms, are stable. Protons are about 1,836 times heavier than electrons, and neutrons are about 1,838 times heavier than electrons. The energy balance in the decay of a neutron is achieved by the anti-neutrino, a neutral particle that carries off surplus energy as the neutron decays. The nominal mass of an atom of an element is measured by the sum of the protons and neutrons in it. This integer is called the **mass number.** The nominal mass of an atom is not affected by the number of electrons, which are very light. Hence the nominal mass, based on the mass number, *approximates* the actual atomic mass. The number of protons in the nucleus, which determines the chemical properties of an element, is called the **atomic number.** Elements are arranged in ascending order of atomic number in an arrangement called the periodic table. The term derives from the tendency to periodicity of chemical properties deriving from arrangements of electrons in atoms.

Radioactive Decay

The nuclei of some elements are not stable. These nuclei are **radioactive,** in that they emit energy and particles, collectively called "radiation." All elements have at least some isotopes that are radioactive. All isotopes of heavy elements with mass numbers greater than 206 and atomic numbers greater than 83 are radioactive.

There are several ways in which unstable nuclei undergo radioactive decay:

- Alpha decay, which the emission of a helium-4 nucleus containing two protons and two neutrons. This is the least penetrating form of radiation. It is stopped by the dead layer of skin and so does no harm when outside the body. But it is the most damaging form of radiation when deposited inside the body.

- Beta decay, which the emission of an electron or a positron (a particle identical to an electron except that it has a positive electrical charge).

- Electron capture, which is the capture by the nucleus of an electron from among the ones whirling around it. In effect, the electron combines with a proton to yield a neutron.

- Spontaneous fission, which is the fission of a heavy element without input of any external particle or energy.

Often, there is still excess residual energy in the nucleus after the emission of a particle or after electron capture. Some of this residual energy after radioactive decay can be emitted in the form of high-frequency electromagnetic radiation, called gamma rays. Gamma rays are essentially like X-rays and are the most penetrating form of radiation.[1] It should be noted that the emission of gamma rays does not change the mass number or atomic number of the nucleus -- that is, unlike radioactive decay by emission of particles, spontaneous fission, or electron capture, it does not cause the transmutation of the nucleus into another element.

Each quantum, or unit, of a gamma ray (or other electromagnetic energy) is called a **photon.** Gamma rays are like light, except that they are much higher frequency electromagnetic rays. Photon energy is directly proportional to the frequency of the electromagnetic radiation. Photons of gamma rays can damage living cells by splitting molecules apart or ionizing elements in them.

Many heavy nuclei emit an energetic alpha particle when they decay. For instance uranium-238 decays into thorium-234 with a half-life of almost 4.5 billion years by emitting an alpha particle:

92-uranium-238 ====> 90-thorium-234 + alpha particle (nucleus of 2-helium-4)

The mass number of uranium-238 declines by four and its atomic number by two when it emits an alpha particle. The number before the element name is the atomic number and that after the element name is the mass number. The totals of the atomic numbers and the mass numbers, respectively, on both sides of the nuclear reaction must be the same. (This is like balancing a chemical equation, in which the number of atoms of each element on both sides of the reaction must be equal)

In beta decay, the atomic number increases by one if an electron is emitted or decreases by one if a positron is emitted. For instance thorium-234, which is the decay product of uranium-238, in turn beta-decays into protactinium-234 by emitting an electron:

90-thorium-234 ====> 91-protactinium-234 + beta particle (electron)

The nuclei that result from radioactive decay may themselves be radioactive. Therefore, some radioactive elements have decay chains that may contain many radioactive elements, one derived from the other. (See Appendix B for a diagram of the decay chain of uranium-238.)

The radioactive decay of nuclei is described probabilistically. Within any given time period, a particular unstable nucleus has a fixed probability of decay. As a result, each radioactive element is characterized by a "half-life," which is the time it takes for half the initial atoms to decay (or transmute into another element or nuclear state). At the end of one half-life, half the original element is left, while the other half is transformed into another element. After two half-lives, one fourth of the original element is left; after three half-lives one eighth is left, and so on. This results in the build-up of decay products. If the decay products themselves decay into other elements, a whole host of radioactive materials come into being. The decay products of radioactive elements are also called **daughter products** or **progeny.**

Binding Energy

Nuclei are tightly bound together by the strong nuclear force and each nucleus has a characteristic **binding energy.** This is the amount of energy it would take to completely break up a nucleus and separate all the neutrons and protons in it. Typically, binding energy increases by several megaelectron-volts (MeV) for every proton or neutron added to a nucleus. (Since protons and neutrons are constituent particles of nuclei, they are known collectively as **nucleons.**) The release of nuclear energy derives from the differences in binding energy between the initial nucleus (or nuclei) and relative to the end-products of the nuclear reaction, such as fission or fusion.

The electrons that whirl around the nucleus are held together in their orbits by electrical forces. It takes on the order of a few electron-volts to dislodge an electron from the outer shell of an atom. The "binding energy" of a nucleon is on the order of a million times greater. Electrons are the particles the enable chemical reactions; nucleons take part in nuclear reactions. The huge differences in binding energy are one measure of the differences in the quantities of energy derived from nuclear compared to chemical reactions.

It must be stressed that the binding energy is the amount of *energy that would have to be added to the nucleus to break it up.* It can be thought of (approximately) as the amount of energy liberated when a nucleon is drawn into the nucleus due to the short range nuclear attractive force. Since energy and mass are equivalent, *nuclei with higher binding energy per nucleon have a lower atomic weight per nucleon.*

The key to release of nuclear energy from fission of heavy elements and fusion of light elements is that elements in the middle of the periodic table of elements, with intermediate mass numbers have a higher binding energy per nucleon (that is a lower atomic weight per nucleon). Therefore when a heavy nucleus is fissioned, the resultant products of the nuclear reaction have a slightly smaller combined nuclear mass. This mass difference is converted to energy during nuclear fission.

Nuclear Fission

Nuclear energy is produced by the conversion of a small amount of the mass of the nucleus of an atom into energy. In principle, all mass and energy are equivalent in a proportion defined by Albert Einstein's famous equation

$E = mc^2$

where E stands for energy, m for mass and c for the speed of light. Since the speed of light is a very large number--300 million meters per second--a small amount of mass is equivalent to a very large amount of energy. For instance, one kilogram (about 2.2 pounds) of matter is equivalent to

$$E = 1 \text{ kg} \times (3 \times 10^8 \text{ meters/sec})^2$$
$$= 1 \times 3 \times 10^8 \times 3 \times 10^8 \text{ joules}$$
$$= 9 \times 10^{16} \text{ joules}$$

This is a huge of amount of energy, equivalent to the energy content of over three million metric tons of coal.

Heavy atoms such as uranium or plutonium can be split by bombarding them with neutrons.[2] The resultant fragments, called fission products, are of intermediate atomic weight, and have a combined mass that is slightly smaller than the original nucleus. The difference appears as energy. As explained in the previous section, this mass difference arises from the binding energy characteristics of heavy elements compared to elements of intermediate atomic weight. Since the binding energy of the fission products per nucleon is higher, their total nucleonic mass is lower. The net result is that fission converts some of the mass of the heavy nucleus into energy.

The energy and mass aspects of the fission process can be explained mathematically as follows. Let the total binding energy of the heavy nucleus and the two fission products be B_h, B_{f1}, and B_{f2}, respectively. Then:

Amount of energy released per fission = $E_r = (B_{f1} + B_{f2}) - B_h$

Amount of mass converted to energy = $Er/c^2 = \{(B_{f1} + B_{f2}) - B_h\}/ c^2$

This energy appears in various forms: the kinetic energy of the neutrons, the vibrational energy of the fission fragments, and gamma radiation. All of these forms of energy are converted to heat by absorption in with the surrounding media in the reactor, mainly the coolant and the moderator (for thermal reactors). The most basic fission reaction in nuclear reactors involves the splitting of the nucleus of uranium-235 when it is struck by a neutron. The uranium-235 first absorbs the neutron to yield uranium-236, and most of these U-236 nuclei split into two fission fragments. Fission reactions typically also release two to four neutrons (depending on the speed on the neutrons inducing the fission and probabilistic factors). One of these neutrons must trigger another fission for a sustained chain reaction. The fission reactions in a nuclear reactor can be written generically as follows:

U-235 + n ==> U-236

U-236 ===> fission fragments + 2 to 4 neutrons + 200 MeV energy (approx.)

The uranium-236 nucleus does not split evenly into equal fission fragments. Rather, the tendency, especially with fission induced by thermal neutrons, is for one fragment to be considerably lighter than the other. Figure 9 (not available in on-line version of report) shows the distribution of fission products due to fission with the slow neutrons and fast neutrons. It can be seen that the fission product atomic numbers are concentrated in the ranges from about 80 to 105 and from about 130 to 150 in thermal reactors. An example of a fission reaction is:

92-U-235 + n ==> 92-U-236

92-U-236 ===> 38-strontium-90 + 54-xenon-144 + 2 neutrons + energy

While many heavy nuclei can be fissioned with fast neutrons, only a few can be fissioned with "slow" neutrons. It turns out that, with some exceptions, like plutonium-240, only nuclei that can be fissioned with slow neutrons can be used for sustaining chain reactions.

Figure A-1: Distribution of Atomic Numbers of Fission Products

[For figure, see report hard copy or p. 213 of *Nuclear Power Deception*]

Source: Till and Meyers, eds. 1983, p. 1-5.

Isotopes with nuclei that can be fissioned with zero energy neutrons (in practice neutrons with low energy, or "slow neutrons") are called *fissile* materials. Generally these are the odd-numbered isotopes, such as uranium-233, uranium-235, plutonium-239, and plutonium-241. Other heavy nuclei, like uranium-238, can be fissioned with fast neutrons, and so are *fissionable*, but not fissile.

There are only three fissile isotopes of practical importance: uranium-233, uranium-235, and plutonium-239. Of these, only uranium-235 occurs naturally in significant quantities. The other two occur in trace quantities only.

Fertile Materials

To obtain plutonium-239 and uranium-233 in amounts useful for nuclear energy production, they must be manufactured from materials that occur in relative abundance. Plutonium-239 is produced from reactions following the absorption of a neutron by uranium-238; uranium-233 is produced by neutron absorption in thorium-232. Uranium-238 and thorium-232 are called **fertile materials,** and the production of fissile materials from them is called **breeding.** The reactions for plutonium-239 are

92-U-238 + n ===> 92-U-239

92-U-239 ====> 93-Np-239 + beta particle (electron)

93-Np-239 ====> 94-Pu-239 + beta particle (electron)

For uranium-233 the reactions are:

90-Th-232 + n ===> 90-Th-233

90-Th-233 ===> 91-Pa-233 + beta particle (electron)

91-Pa-233 ====> 92-U-233 + beta particle.

The symbol Pa stands for the element protactinium.

Endnotes to Appendix A:

1. The terms alpha, beta, and gamma radiation, and X-rays were coined because scientists did not know the nature of these kinds of radiation when they were first detected.

2. Nuclear fission can also be induced by bombardment of the nucleus by electrically charged particles, such as alpha particles. However, the nucleus is positively charged and alpha particles are also positively charged. Since positive charges repel each other, these types of fission reactions are more difficult to accomplish than reactions with neutrons. Fission can also be induced by bombarding the nucleus with energetic gamma rays (photons). This process is called photofission.

Appendix B: Plutonium and Uranium

Reprinted from *Physical, Nuclear, and Chemical, Properties of Plutonium*, IEER fact sheet, February 1997, on the Web at www.ieer.org/fctsheet/pu-props.html, and from *Uranium: Its Uses and Hazards*, IEER fact sheet, updated August 24, 2000, on the Web at www.ieer.org/fctsheet/uranium.html.

[Appendix B in hard copy versions of *Nuclear Alchemy Gamble* consisted of material reprinted from pp. 215-227 of *Nuclear Power Deception: U.S. Nuclear Mythology from Electricity "Too Cheap to Meter" to "Inherently Safe" Reactors*, by Arjun Makhijani and Scott Saleska, Apex Press, 1999. This material is very similar to the fact sheets from IEER's Web site reprinted here.]

Physical, Nuclear, and Chemical, Properties of Plutonium

Plutonium-239 is one of the two fissile materials used for the production of nuclear weapons and in some nuclear reactors as a source of energy. The other fissile material is uranium-235. Plutonium-239 is virtually nonexistent in nature. It is made by bombarding uranium-238 with neutrons in a nuclear reactor. Uranium-238 is present in quantity in most reactor fuel; hence plutonium-239 is continuously made in these reactors. Since plutonium-239 can itself be split by neutrons to release energy, plutonium-239 provides a portion of the energy generation in a nuclear reactor.

The physical properties of plutonium metal are summarized in Table 1.

TABLE 1. Physical Characteristics of Plutonium Metal

Color:	silver
Melting point:	641 deg. C
Boiling point:	3232 deg. C
Density:	16 to 20 grams/cubic centimeter

Nuclear Properties of Plutonium

Plutonium belongs to the class of elements called transuranic elements whose **atomic number** is higher than 92, the atomic number of uranium. Essentially all transuranic materials in existence are manmade. The atomic number of plutonium is 94.

Plutonium has 15 isotopes with **mass numbers** ranging from 232 to 246. Isotopes of the same element have the same number of protons in their nuclei but differ by the number of neutrons. Since the chemical characteristics of an element are governed by the number of protons in the nucleus, which equals the number of electrons when the atom is electrically neutral (the usual elemental form at room temperature), all isotopes have nearly the same chemical characteristics. This means that in most cases it is very difficult to separate isotopes from each other by chemical techniques.

Only two plutonium isotopes have commercial and military applications. Plutonium-238, which is made in nuclear reactors from neptunium-237, is used to make compact thermoelectric generators; plutonium-239 is used for nuclear weapons and for energy; plutonium-241, although fissile, (see next paragraph) is impractical both as a nuclear fuel and a material for nuclear warheads. Some of the reasons are far higher cost , shorter half-life, and higher radioactivity than plutonium-239. Isotopes of plutonium with mass numbers 240 through 242 are made along with plutonium-239 in nuclear reactors, but they are contaminants with no commercial applications. In this fact sheet we focus on civilian and military plutonium (which are interchangeable in practice--see Table 5), which consist mainly of plutonium-239 mixed with varying amounts of other isotopes, notably

plutonium-240, -241, and -242.

Plutonium-239 and plutonium-241 are fissile materials. This means that they can be split by both slow (ideally zero-energy) and fast neutrons into two new nuclei (with the concomitant release of energy) and more neutrons. Each fission of plutonium-239 resulting from a slow neutron absorption results in the production of a little more than two neutrons on the average. If at least one of these neutrons, on average, splits another plutonium nucleus, a sustained chain reaction is achieved.

The even isotopes, plutonium-238, -240, and -242 are not fissile but yet are fissionable--that is, they can only be split by high energy neutrons. Generally, fissionable but non-fissile isotopes cannot sustain chain reactions; plutonium-240 is an exception to that rule.

The minimum amount of material necessary to sustain a chain reaction is called the critical mass. A supercritical mass is bigger than a critical mass, and is capable of achieving a growing chain reaction where the amount of energy released increases with time.

The amount of material necessary to achieve a critical mass depends on the geometry and the density of the material, among other factors. The critical mass of a bare sphere of plutonium-239 metal is about 10 kilograms. It can be considerably lowered in various ways.

The amount of plutonium used in fission weapons is in the 3 to 5 kilograms range. According to a recent Natural Resources Defense Council report(1), nuclear weapons with a destructive power of 1 kiloton can be built with as little as 1 kilogram of weapon grade plutonium(2). The smallest theoretical critical mass of plutonium-239 is only a few hundred grams.

In contrast to nuclear weapons, nuclear reactors are designed to release energy in a sustained fashion over a long period of time. This means that the chain reaction must be controlled--that is, the number of neutrons produced needs to equal the number of neutrons absorbed. This balance is achieved by ensuring that each fission produces exactly one other fission.

All isotopes of plutonium are radioactive, but they have widely varying half-lives. The half-life is the time it takes for half the atoms of an element to decay. For instance, plutonium-239 has a half-life of 24, 110 years while plutonium-241 has a half-life of 14.4 years. The various isotopes also have different principal decay modes. The isotopes present in commercial or military plutonium-239 are plutonium-240, -241, and -242. Table 2 shows a summary of the radiological properties of five plutonium isotopes.

The isotopes of plutonium that are relevant to the nuclear and commercial industries decay by the emission of alpha particles, beta particles, or **spontaneous fission. Gamma radiation**, which is penetrating electromagnetic radiation, is often associated with **alpha and beta decays**.

TABLE 2. Radiological Properties of Important Plutonium Isotopes

	Pu-238	**Pu-239**	**Pu-240**	**Pu-241**	**Pu-242**
Half-life(in years)	87.74	24,110	6537	14.4	376,000
Specific activity(curies/gram)	17.3	.063	.23	104	.004
Principal decay mode	alpha	alpha	alpha some spontaneous fission(a)	beta	alpha
Decay energy(MeV)	5.593	5.244	5.255	.021	4.983
Radiological hazards	alpha, weak gamma	alpha, weak gamma	alpha, weak gamma	beta, weak gamma(b)	alpha, weak gamma

Source: CRC Handbook of Chemistry and Physics; 1990-1991. Various sources give slightly different figures for half-lives and energies.
a) Source of neutrons causing added radiation dose to workers in nuclear facilities. A little spontaneous fission occurs in most plutonium isotopes.
b) Plutonium-241 decays into Americium-241, which is an intense gamma-emitter.

Chemical properties and hazards of plutonium.

Table 3 describes the chemical properties of plutonium in air. These properties are important because they affect the safety of storage and of operation during processing of plutonium. The oxidation of plutonium represents a health hazard since the resulting stable compound, plutonium dioxide is in particulate form that can be easily inhaled. It tends to stay in the lungs for long periods, and is also transported to other parts of the body. Ingestion of plutonium is considerably less dangerous since very little is absorbed while the rest passes through the digestive system.

TABLE 3. How Plutonium Metal Reacts in Air

Forms and Ambient Conditions:	*Reaction:*
Non-divided metal at room temperature (corrodes)	relatively inert, slowly oxidizes
Divided metal at room temperature (PuO2)	readily reacts to form plutonium dioxide
Finely divided particles under about 1 millimeter diameter particles over about 1 millimeter diameter	spontaneously ignites at about 150 C(c) spontaneously ignites at about 500 C.
Humid, elevated temperatures (PuO2)	readily reacts to form plutonium dioxide

c) US Department of Energy, "Assessment of Plutonium Storage Safety Issues at DOE Facilities," DOE/DP-0123T (Washington, DC: US DOE, Jan 1994.

Important Plutonium Compounds and their Uses

Plutonium combines with oxygen, carbon, and fluorine to form compounds which are used in the nuclear industry, either directly or as intermediates.

Table 4 shows some important plutonium compounds. Plutonium metal is insoluble in nitric acid and plutonium is slightly soluble in hot, concentrated nitric acid. However, when plutonium dioxide and uranium dioxide form a solid mixture, as in spent fuel from nuclear reactors, then the solubility of plutonium dioxide in nitric acid is enhanced due to the fact that uranium dioxide is soluble in nitric acid. This property is used when **reprocessing** irradiated nuclear fuels.

TABLE 4. Important Plutonium Compounds and Their Uses

Compound:	*Use:*
Oxides Plutonium Dioxide(PuO2)	can be mixed with uranium dioxide (UO2) for use as reactor fuel
Carbides Plutonium Carbide(PuC) Plutonium Dicarbide(PuC2) Diplutonium Tricarbide(Pu2C3)	all three carbides can potentially be used as fuel in breeder reactors
Fluorides Plutonium Trifluoride(PuF3) Plutonium Tetrafluoride(PuF4)	both fluorides are intermediate compounds in the production of plutonium metal
Nitrates Plutonium Nitrates [Pu(NO3)4] and [Pu(NO3)3]	no use, but it is a product of reprocessing (extraction of plutonium from used nuclear fuel).

Formation and Grades of Plutonium-239

Plutonium-239 is formed in both civilian and military reactors from uranium-238.

The subsequent absorption of a neutron by plutonium-239 results in the formation of plutonium-240. Absorption of another neutron by plutonium-240 yields plutonium-241. The higher isotopes are formed in the same way. Since plutonium-239 is the first in a string of plutonium isotopes created from uranium-238 in a reactor, the longer a sample of uranium-238 is irradiated, the greater the percentage of heavier isotopes. Plutonium must be chemically separated from the fission products and remaining uranium in the irradiated reactor fuel. This chemical separation is called reprocessing.

high "burn-up", because it is fuel irradiation that generates the heat required for power production. If the goal is production of plutonium for military purposes then the "burn-up" is kept low so that the plutonium-239 produced is as pure as possible, that is, the formationo of the higher isotopes, particularly plutonium-240, is kept to a minimum.

Plutonium has been classified into grades by the US DOE (Department of Energy) as shown in Table 5.

It is important to remember that this classification of plutonium according to grades is somewhat arbitrary. For example, although "fuel grade" and "reactor grade" are less suitable as weapons material than "weapon grade" plutonium, they can also be made into a nuclear weapon, although the yields are less predictable because of unwanted neutrons from spontaneous fission. The ability of countries to build nuclear arsenals from reactor grade plutonium is not just a theoretical construct. It is a proven fact. During a June 27, 1994 press conference, Secretary of Energy Hazel O'Leary revealed that in 1962 the United States conducted a successful test with "reactor grade" plutonium. All grades of plutonium can be used as weapons of radiological warfare which involve weapons that disperse radioactivity without a nuclear explosion.

TABLE 5. Grades of Plutonium

Grades	Pu-240 Content
Supergrade	2-3 %
Weapon grade	< 7 %
Fuel grade	7-19 %
Reactor grade	19 % or greater

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Uranium: Its Uses and Hazards

First discovered in the 18th century, uranium is an element found everywhere on Earth, but mainly in trace quantities. In 1938, German physicists Otto Hahn and Fritz Strassmann showed that uranium could be split into parts to yield energy. Uranium is the principal fuel for nuclear reactors and the main raw material for nuclear weapons.

Natural uranium consists of three isotopes: uranium-238, uranium-235, and uranium-234. Uranium isotopes are radioactive. The nuclei of radioactive elements are unstable, meaning they are transformed into other elements, typically by emitting particles (and sometimes by absorbing particles). This process, known as radioactive decay, generally results in the emission of alpha or beta particles from the nucleus. It is often also accompanied by emission of gamma radiation, which is electromagnetic radiation, like X-rays. These three kinds of radiation have very different properties in some respects but are all ionizing radiation--each is energetic enough to break chemical bonds, thereby possessing the ability to damage or destroy living cells.

Summary of Uranium Isotopes				
Isotope	**Percent in natural uranium**	**No. of Protons**	**No. of Neutrons**	**Half-Life (in years)**
Uranium-238	99.284	92	146	4.46 billion
Uranium-235	0.711	92	143	704 million
Uranium-234	0.0055	92	142	245,000

Uranium-238, the most prevalent isotope in uranium ore, has a half-life of about 4.5 billion years; that is, half the atoms in any sample will decay in that amount of time. Uranium-238 decays by alpha emission into thorium-234, which itself decays by beta emission to protactinium-234, which decays by beta emission to uranium-234, and so on. The various decay products, (sometimes referred to as "progeny" or "daughters") form a series starting at uranium-238. After several more alpha and beta decays, the series ends with the stable isotope lead-206.

URANIUM DECAY CHAIN -- Main Branch Read from left to right. Arrows indicate decay.		
Uranium-238 ==> (half-life: 4.46 billion years) alpha decay	Thorium-234 ==> (half-life: 24.1 days) beta decay	Protactinium-234m ==> (half-life: 1.17 minutes) beta decay
Uranium-234 ==> (half-life: 245,000 years) alpha decay	Thorium-230 ==> (half-life: 75,400 years) alpha decay	Radium-226 ==> (half-life: 1,600 years) alpha decay
Radon-222 ==> (half-life: 3.82 days) alpha decay	Polonium-218 ==> (half-life: 3.11 minutes) alpha decay	Lead-214 ==> (half-life: 26.8 minutes) beta decay
Bismuth-214 ==> (half-life: 19.9 minutes) beta decay	Polonium-214 ==> (half-life: 163 microseconds) alpha decay	Lead-210 ==> (half-life: 22.3 years) beta decay
Bismuth-210 ==> (half-life: 5.01 days) beta decay	Polonium-210 ==> (half-life: 138 days) alpha decay	Lead-206 (stable)

Uranium-238 emits alpha particles which are less penetrating than other forms of radiation, and weak gamma rays As long as it remains outside the body, uranium poses little health hazard (mainly from the gamma-rays). If inhaled or ingested, however, its radioactivity poses increased risks of lung cancer and bone cancer. Uranium is also chemically toxic at high concentrations and can cause damage to internal organs, notably the kidneys. Animal studies suggest that uranium may affect reproduction, the developing fetus,[1] and increase the risk of leukemia and soft tissue cancers.[2]

The property of uranium important for nuclear weapons and nuclear power is its ability to fission, or split into two lighter fragments when bombarded with neutrons releasing energy in the process. Of the naturally-occurring uranium isotopes, only uranium-235 can sustain a chain reaction-- a reaction in which each fission produces enough neutrons to trigger another, so that the fission process is maintained without any external source of neutrons.[3] In contrast, uranium-238 cannot sustain a chain reaction, but it can be converted to plutonium-239, which can.[4] Plutonium-239, virtually non-existent in nature, was used in the first atomic bomb tested July 16, 1945 and the one dropped on Nagasaki on August 9, 1945.

The Mining and Milling Process

Traditionally, uranium has been extracted from open-pits and underground mines. In the past decade, alternative techniques such in-situ leach mining, in which solutions are injected into underground deposits to dissolve uranium, have become more widely used. Most mines in the U.S. have shut down and imports account for about three-fourths of the roughly 16 metric tons of refined uranium used domestically each year -- Canada being the largest single supplier.[5]

The milling (refining) process extracts uranium oxide (U_3O_8) from ore to form yellowcake, a yellow or brown powder that contains about 90 percent uranium oxide.[6] Conventional mining techniques generate a substantial quantity of mill tailings waste during the milling phase, because the usable portion is generally less than one percent of the ore. (In-situ leach mining leaves the unusable portion in the ground, it does not generate this form of waste). The total volume of mill tailings generated in the U.S. is over 95 percent of the volume of all radioactive waste from all stages of the nuclear weapons and power production.[7] While the hazard per gram of mill tailings is low relative to most other radioactive wastes, the large volume and lack of regulations until 1980 have resulted in widespread environmental contamination. Moreover, the half-lives of the principal radioactive components of mill tailings, thorium-230 and radium-226 are long, being about 75,000 years and 1,600 years respectively.

The most serious health hazard associated with uranium mining is lung cancer due to inhaling uranium decay products. Uranium mill tailings contain radioactive materials, notably radium-226, and heavy metals (e.g., manganese and molybdenum) which can leach into groundwater. Near tailings piles, water samples have shown levels of some contaminants at hundreds of times the government's acceptable level for drinking water.[8]

Mining and milling operations in the U.S. have disproportionately affected indigenous populations around the globe. For example, nearly one third of all mill tailings from abandoned mill operations are on lands of the Navajo nation alone.[9] Many Native Americans have died of lung cancers linked to their work in uranium mines. Others continue to suffer the effects of land and water contamination due to seepage and spills from tailings piles.[10]

Conversion and Enrichment

Uranium is generally used in reactors in the form of uranium dioxide (UO_2) or uranium metal; nuclear weapons use the metallic form. Production of uranium dioxide or metal requires chemical processing of yellowcake. Further, most civilian and many military reactors require uranium that has a higher proportion of uranium-235 than present in natural uranium. The process used to increase the amount of uranium-235 relative to uranium-238 is known as uranium enrichment.

U.S. civilian power plants typically use 3 to 5 percent uranium-235. Weapons use "highly enriched uranium" (HEU) with over 90 percent uranium-235. Some

research reactors and all U.S. naval reactors also use HEU.

To enrich uranium, it must first be put in the chemical form uranium hexafluoride (UF_6). After enrichment, UF6 is chemically converted to uranium dioxide or metal. A major hazard in both the uranium conversion and uranium enrichment processes comes from the handling of uranium hexafluoride, which is chemically toxic as well as radioactive. Moreover, it reacts readily with moisture, releasing highly toxic hydrofluoric acid. Conversion and enrichment facilities have had a number of accidents involving uranium hexafluoride.[11]

The bulk of waste from the enrichment process is depleted uranium--so-called because most of the uranium-235 has been extracted from it. Depleted uranium has been used by the U.S. military to fabricate armor-piercing conventional weapons and tank armor plating. It was incorporated into these conventional weapons without informing armed forces personnel that depleted uranium is a radioactive material and without procedures for measuring doses to operating personnel.

The enrichment process can also be reversed. Highly enriched uranium can be diluted, or "blended down" with depleted, natural, or very low-enriched uranium to produce 3 to 5 percent low-enriched reactor fuel. Uranium metal at various enrichments must be chemically processed so that it can be blended into a homogeneous material at one enrichment level. As a result, the health and environmental risks of blending are similar to those for uranium conversion and enrichment.

Regulations in the U.S.

In 1983 the federal government set standards for controlling pollution from active and abandoned mill tailings piles resulting from yellowcake production. The principal goals of federal regulations are to limit the seepage of radionuclides and heavy metals into groundwater and reduce emissions of radon-222 to the air. Mandatory standards for decommissioning nuclear facilities including conversion and enrichment facilities are only now being developed by the U.S. Environmental Protection Agency and the U.S. Nuclear Regulatory Commission (NRC). So far, the NRC has been using guidelines developed by its staff in 1981 to oversee decommissioning efforts.[12]

The Future

Uranium and associated decay products thorium-230 and radium-226 will remain hazardous for thousands of years. Current U.S. regulations, however, cover a period of 1,000 years for mill tailings and at most 500 years for "low-level" radioactive waste. This means that future generations--far beyond those promised protection by these regulations--will likely face significant risks from uranium mining, milling, and processing activities.

Endnotes

1. Agency for Toxic Substances and Disease Registry, ATSDR Public Health Statement: Uranium, Atlanta: ATSDR, December 1990.

2. Filippova, L. G., A. P. Nifatov, and E. R. Lyubchanskii, Some of the long-term sequelae of giving rats enriched uranium (in Russian), Radiobiologiya, v. 18, n. 3,, pp. 400-405. 1978. Translated in NTIS UB/D/120-03 (DOE-TR-4/9), National Technical Information Service, Springfield, Virginia.

3. Uranium-235 and plutonium-239 are called "fissile" isotopes--defined as materials that can be fissioned by low-energy (ideally zero energy) neutrons.

4. Uranium-238 is converted to plutonium-239 by bombarding it with neutrons: U-238 + neutron -->> U-239 U-239 ==> Np-239 + beta particle (electron) Np-239 ==> Pu-239 + beta particle (electron)

5. Energy Information Administration, Uranium Purchases Report 1992, DOE/EIA-0570(92), Washington, D.C., August 1993. The number of conventional mines operating in the U.S. has declined from a peak of hundreds to zero in 1993, seven "non-conventional" mining operations (e.g., in-situ leach) accounted for all domestic ore production for that year. (NUEXCO, NUEXCO Review: 1993 Annual, Denver, 1994).

6. Benedict, Manson, Thomas Pigford, and Hans Wolfgang Levi. Nuclear Chemical Engineering. 2 ed.. (New York: McGraw-Hill Book Company, 1981), p. 265. Note that pure U3O8 is black. Yellowcake gets its color from the presence of ammonium diuranate.

7. Based on the total volume of all radioactive waste (including spent fuel, high-level waste, transuranic waste, low-level waste and uranium mill tailings) from all sources (both commercial and military) produced in the U.S. since the 1940s, as compiled in Scott Saleska, et al. Nuclear Legacy: An Overview of the Places, Politics, and Problems of Radioactive Waste in the United States (Washington, DC: Public Citizen, 1989), Appendix C.

8. U.S. Environmental Protection Agency, Final Environmental Impact Statement for Standards for the Control of Byproduct Materials from Uranium Ore Processing, Washington, D.C., 1983, v. 1, pp. D-12, D-13.

9. Gilles, Cate, Marti Reed, and Jacques Seronde, Our Uranium Legacy, 1990 [available from Southwest Research and Information Center, Albuquerque, NM].

10. In 1979, a dam holding water in a mill tailings settling pond at the United Nuclear Fuels Corporation mill near Church Rock, New Mexico gave way and released about 100 million gallons of contaminated water into the Puerco River which cuts through Navajo grazing lands.

11. One such accident at the Sequoyah Fuels conversion plant in Gore, Oklahoma killed one worker, hospitalized 42 others, and approximately 100 residents.

12 .For more information about cleanup standards, see Science for Democratic Action, (IEER, Takoma Park, MD), Vol. 3, No. 1, Winter 1994.

Appendix C: Basics of Nuclear Reactors

Reprinted from *Nuclear Power Deception: U.S. Nuclear Mythology from Electricity "Too Cheap to Meter" to "Inherently Safe" Reactors,* by Arjun Makhijani and Scott Saleska, Apex Press, 1999, pp. 36-52.

Appendix C: Basics of Nuclear Reactors

Nuclear Reactors

Nuclear power plants, it should be clear, are complex installations and by their nature, they must be designed with care.

--John R. Lamarsh, Introduction to Nuclear Engineering, a textbook[28]

As we have discussed, energy from nuclear fission comes from the transformation into energy of a small amount of the mass of a heavy nucleus when it is split. When the nucleus of uranium-235 or plutonium-239 is fissioned, the resulting energy takes many forms. Some of the energy is released in the form of high speed neutrons, some appears as electromagnetic radiation (gamma rays); most is released as vibrational energy of the fission fragments. Almost all this energy is quickly transformed into thermal energy, or heat. A nuclear reactor is basically a vessel that is designed to capture this heat energy in a liquid or gas medium called a coolant in a sustained and controlled way. A nuclear reactor must have the following features:

- It must accommodate a sufficient number of fuel rods to sustain a chain reaction at the maximum level of thermal power to be generated. (Power is defined as the rate of energy production).
- It must incorporate ways to control the chain reaction, so that the level of power output can be maintained constant at the required level or varied from zero to the maximum, as necessary, without the danger of severe runaway nuclear reactions.
- There must be ways to capture the energy from the fission reactions and radioactive decay of the fission products and transport it out of the reactor vessel.
- The vessel must be strong enough to withstand high temperatures and (in most cases) high pressures, as well as intense neutron bombardment.
- The vessel and the structure in which it is located must contain the radiation within them so far as possible to minimize radiation doses to workers and off-site populations.

The central function of the nuclear reactor is to generate heat at the required rate in order to drive a heat engine. A number of different reactors have been designed to accomplish this. Another function of reactors is to convert uranium-238 into plutonium-239, though in most commercial reactors this has become a secondary function. In fact, in the context of non-proliferation, it is a problem. Reactors designed specifically to produce more fissile material than they consume as a result of the conversion of uranium-238 into fissile plutonium isotopes are called "breeder reactors."[29]

Reactors are classified into two types: *thermal reactors,* which use thermal (or "slow") neutrons to sustain the chain reaction, and *fast reactors,* which use fast, or energetic, neutrons to sustain the chain reaction.

1. Thermal reactors

The design of nuclear reactors depends centrally on the type of coolant that is used to carry off the heat produced in the reactor vessel. For thermal reactors, it also depends on

the choice of a material called the *moderator*, which slows down the fast neutrons emitted in the process of fission.

Sustained chain reactions can be achieved with smaller proportions of fissile isotopes in the reactor fuel if the neutrons emitted from fission reactions are slowed down. For instance, some reactors that use slow neutrons can even use natural uranium as a fuel, even though it contains only about 0.7 percent of fissile uranium-235. Slow neutrons, called thermal neutrons, have energies of a fraction of an electron-volt (eV). Neutrons from fission reactions typically have energies of several megaelectron-volts (MeV) at the time they are emitted.

The process of slowing down neutrons in a nuclear reactor is called *moderation.* It is achieved by putting a moderator in a nuclear reactor. A moderator should preferentially be a light element so that neutrons can slow down when they collide with its atoms. For the most part, this happens by elastic collisions. This process is analogous to that by which billiard balls slow down when they collide with balls of similar weight. Heavy atoms would make less suitable moderators since neutrons would not lose as much energy to them in collisions. This can be visualized as billiard balls simply bouncing off when they collide with the (far heavier) edge of the pool table. Many collisions are needed to slow down fast neutrons to thermal energies. These collisions convert the kinetic energy of the fast neutrons into heat, which is randomized rather than directed kinetic energy. Finally, the moderator must also not absorb too many neutrons in the process of slowing them down. Otherwise sufficient neutrons will not remain to sustain a chain reaction.

Transfer of energy out of the reactor vessel requires that a coolant flow through it. Without a coolant, continued production of fission energy would cause the reactor vessel and its contents to get very hot. This would rapidly lead to a melting of the fuel and fuel rods, a phenomenon called a "meltdown." The coolant must also carry away the heat generated by the radioactive decay of fission products, which build up in the reactor as the fission process continues. When a reactor has been operating for a long-time, the heat from decaying fission products alone amounts to several percent of the full power rating. Loss of coolant in a reactor can produce a meltdown in such cases just due to the failure to carry away the decay heat from the fission products. For instance, this was the cause of the partial meltdown in Three Mile Island Unit 2 in 1979.[30]

In some reactors, the coolant and moderator are the same material. Hydrogen is an excellent moderator, being light and having a low neutron absorption cross-section (or probability). However, hydrogen gas is explosive and so it is used in the chemical form of ordinary water, H2O, also called light water. Further, the density of hydrogen in water (that is, the number of hydrogen atoms per unit volume of water) is far greater than that of hydrogen gas. Thus, a smaller volume of water gives the same amount of moderation as a far greater volume of hydrogen gas. Besides working well as a moderator, water is also a good coolant. Thus, the most common reactor types in the world use light water as a coolant and moderator. They are called light water reactors or LWRs.

Figure 3 [not available in Web version of report] shows a schematic diagram of one type of light water reactor called a boiling water reactor, called a BWR. In these reactors, developed by General Electric, the water that serves as a coolant and moderator in the

reactor is boiled directly in the reactor. This steam is used to drive a turbine. The main advantage of the BWR design is that it does not require an expensive boiler apart from the reactor. There are a number of disadvantages however, including higher emissions of radioactive gases and the fact that the turbines are exposed to radioactive steam.

Light water reactors are also used in another design, called a pressurized water reactor (PWR). This design, which is the most common power reactor design today, has two water circuits. The primary circuit is the high pressure water in the reactor vessel. This water is kept under such high pressure that it does not boil. The hot, high pressure water is passed though a heat exchanger, called a steam generator, where it heats up water in the secondary circuit and converts it into steam, much as the hot gases in a conventional boiler convert water in a boiler into steam. There are usually three or four steam generators in a PWR. The steam generators add considerable expense to the nuclear reactor but keep the radioactive primary coolant out of the turbines. The line diagram of a nuclear power station in Figure 1 above shows a power plant with a steam generator. That figure differs from a PWR only in that it indicates a solid moderator, whereas in a PWR the coolant and moderator are the same -- ordinary water.

Deuterium, or heavy hydrogen (symbol: D), whose nucleus consists of one proton and one neutron, can also be used as a moderator. It is the best moderating material from the point of view of low neutron absorption. Like ordinary hydrogen gas, it is explosive and so is used in the chemical form of water, called heavy water (symbol: D_2O). In contrast to LWRs, heavy water moderated reactors (HWRs) can use natural uranium as fuel. Figure 4 [not available in on-line version of report] shows a diagram of an HWR used for power generation in Canada, called a CANDU (CANada Deuterium Uranium) reactor.

Carbon in the form of graphite is also a good moderator, but carbon-moderated reactors need a separate coolant. The most common coolants are helium gas, carbon dioxide gas, or water. Reactors of the Chernobyl design (called RBMK reactors) use carbon in the form of graphite as a moderator and water as a coolant.

It is also necessary to control the chain reaction in order to vary the power output of the reactor. To maintain power at a sustained fixed level each fission of a heavy nucleus must produce exactly one more fission. This means that only one of the neutrons arising from fission must give rise to another fission. The ratio of the number of fissions that each fission reaction gives rise to (on average) is called the *multiplication factor*. For a sustained power level, the multiplication factor must be precisely equal to one. At this point, the reactor is *critical* and the nuclear chain reaction will sustain itself at constant power output. If the multiplication factor falls below one, the reactor becomes *subcritical* and the chain reaction will stop. If it rises above one, the reactor is *supercritical* and the power level will increase.

A parameter, called *reactivity,* is often used to describe reactor control. It is related to the multiplication factor in the following way: If the multiplication factor is exactly one, the reactivity is exactly zero; if the multiplication factor is greater than one, the reactivity is positive (but less than one). If the multiplication factor is between zero and one, the reactivity is negative. Reactivity is a convenient way to describe reactor control because positive reactivity means a supercritical reactor, zero reactivity means a critical reactor, and negative reactivity means a subcritical reactor.

Start-up, shut down, or change in power level -- that is, control -- of a reactor is accomplished by changing the reactivity.[31] This is done by controlling the number of nuclear fission reactions per second that typically occur in a reactor. A neutron-absorbing material, like boron, is made into rods ("control rods") which are interspersed with the fuel rods and which can be inserted into or removed from the reactor core.[32] This controls the number of neutrons available for fission reactions and the rate of energy production (or power output). A nuclear reactor can be shut down by making the reactivity negative. This is accomplished by inserting the control rods into the reactor far enough so that they will absorb the quantity of neutrons needed to stop the chain reaction. Raising the control rods temporarily makes the reactivity positive, that is, it makes reactor slightly supercritical for a short period of time, enabling an increase in the power level. The reactor is returned to the critical state (reactivity equal to zero) when the desired level of power is achieved.

Control of a reactor can be lost if the reactor continues to stay supercritical (that is, if the reactivity stays positive) for longer than intended. An increase of the multiplication factor is also called a *reactivity insertion.* The intense heat generated by excess fission could overwhelm the cooling systems, causing a severe accident. The most severe accident in nuclear power history, which occurred in reactor number 4 at the Chernobyl power plant on April 26, 1986, involved a loss of control of the nuclear chain reaction.

The time in which reactor power level increases by a factor of about 2.7 (or more accurately, by a factor equal to *e*, the base of natural logarithms) is called the *reactor period.* This quantity depends on the design of the reactor and the composition of the fuel. Power reactors are designed to have long reactor periods in order have slow, smooth increases and decreases in reactor temperature. This minimizes thermal stresses and allows for longer reactor operating lifetime. A typical reactor period in a power reactor would be on the order of one hour.

Control of the reactor is facilitated by the fact that while most (generally more than 99 percent) neutrons from the fission process are emitted essentially at the same time as the fission occurs, a small proportion are emitted after a relatively long time. The former are called prompt neutrons, while the latter are called delayed neutrons. If a reactor becomes critical with only prompt neutrons, the reactor period would be only a tiny fraction of a second, so that control of the reactor would be essentially impossible. But if the reactor is designed so that it does not become critical with prompt neutrons only, then the reactor period and the time available to control it can be increased greatly.

But accidental "prompt criticality" remains a safety concern, since control of the reactor could be lost if a reactor becomes critical with prompt neutrons only. The proportion of delayed neutrons in an LWR is about 0.0065 (that is about two-thirds of one percent).[33] So long as the reactivity of the reactor stays below the proportion of delayed neutrons, the reactor cannot become prompt critical, and can be controlled. An increase of reactivity above the delayed neutron fraction results in the loss of control of the reactor. For comparison, fast neutron reactors using uranium-233 or plutonium-239 fuel are even more difficult to control, since the delayed neutron fraction is only about 0.0020.

Reactors such as LWRs in which fuel is loaded in batches require more complex systems to ensure control because when the fuel is fresh, reactivity increase can be large for a

modest movement of control rods. During such periods, reactor control is enhanced by adding neutron absorbing chemicals to the water. As noted above, this is known as chemical shim.

The ejection of control rods from a reactor that has relatively fresh fuel in it could result in a total loss of reactor control. This is more of a potential problem with batch-fueled reactors, such as LWRs, than with continuous fueled reactors, such as the Canadian heavy water reactor (CANDU).

Commercial light water reactors use uranium fuel enriched to between 3 and 5 percent as a fuel. Graphite or heavy water moderated reactors can use natural uranium as a fuel. This is a considerable advantage in countries that do not have uranium enrichment plants. It was a principal factor that led a number of countries, including the Soviet Union, France, and Britain, to choose graphite-moderated reactors when they began their military plutonium production. U.S. naval reactors use highly enriched uranium (up to 97.6 percent enrichment) as a fuel because this enables the reactors to operate for longer periods without refueling.

Table 2 shows various types of thermal reactors, along with the coolants, moderators, and fuel types they use.

Table 2: Basic Characteristics of Reactors Types

Reactor Type	Light Water Reactor (LWR)		Heavy Water Reactor (HWR)
	a. Boiling Water Reactor	b. Pressurized Water Reactor (PWR)	
Purpose[1]	electricity	electricity; nuclear powered ships (U.S.)	electricity; plutonium production
Coolant Type	water (H_2O)	water	heavy water (deuterium oxide, D_2O)
Moderator Type	water	water	heavy water
Fuel -- Chemical Composition[2]	uranium-dioxide (UO2)	uranium-dioxide	uranium-dioxide or metal
Fuel - Enrichment Level[3]	low-enriched	low-enriched	natural uranium (not enriched)
Comments	steam generated inside the reactor goes directly to the turbine	steam is generated outside the reactor in a secondary heat transfer loop	used in Canada: called "CANDU" - "Canadian Deuterium Uranium;" Also used in Savannah River Site reactors (metal fuel at SRS)

Reactor Type	Graphite Moderated Reactor		Fast Breeder Reactor (FBR) Liquid Metal (LMFBR) (most common type of breeder)
	a. Gas Cooled	b. Water Cooled	
Purpose[1]	electricity; plutonium production	electricity; plutonium production	electricity; plutonium production
Coolant Type	gas (carbon dioxide or helium)	water	molten, liquid sodium
Moderator Type	graphite	graphite	not required
Fuel -- Chemical Composition[2]	uranium dicarbide (UC_2) or uranium metal	uranium dioxide (RBMK) or metal (N-reactor)	plutonium dioxide and uranium dioxide in various arrangements
Fuel - Enrichment Level[3]	slightly-enriched, natural uranium	slightly-enriched	various mixtures of plutonium-239 and uranium-235
Comments	used in Britain, and France (e.g.: AGR, MAGNOX)	used in former Soviet Union, e.g. Chernobyl (RBMK); N-reactor at Hanford.	breeder reactors are designed to produce more fissile material than they consume. Monju; Phenix

Source: Lamarsh, John, *Introduction to Nuclear Engineering,* (Reading, MA: Addison-Wesley publishing Co., 1983), 120-143.

Table notes:

1. The purpose of the reactor does not depend on the choice of coolant or moderator, but rather on reactor size and on how the reactor is operated, and on what ancilliary materials are put into fuel rods besides fuel. The same reactors can, in principle, be used for electricity production, military plutonium production, and production of other radioactive materials such as tritium for military and civilian applications. The purposes listed in this column are the common ones to which such reactors are or have been put.

2. Not all fuel types necessarily included.

3. The enrichment of fuel refers to the percentage of the isotope of uranium-235 compared to uranium-238 present in fuel. It is defined here as follows: slightly enriched uranium = about 0.8 to 3%; low enriched uranium = 3 to 5 %.

2. Breeder Reactors (Fast Neutron Reactors)

As we have discussed above, of the fissile materials usable for practical nuclear energy production, only uranium-235 occurs in any substantial quantities in nature. The other two, plutonium-239 and uranium-233, must be made from uranium-238 and thorium-232 respectively, which are far more abundant than naturally-occurring fissile uranium-235. The process of converting "fertile" uranium-238 and thorium-232 into fissile materials is called "breeding," evidently by analogy with biological reproduction.

Commercial nuclear power reactors use natural or "low-enriched" uranium as fuel. Natural uranium contains 0.711% uranium-235 and "low-enriched" reactor fuel contains from 1% to 5% uranium-235, depending on reactor design. Almost all the rest is uranium-238. (See Appendix B.)

Some of the neutrons in a nuclear reactor convert uranium-238 into plutonium-239. In other words, there is "breeding" of plutonium in all commercial reactors containing uranium-238. However, the term "breeder" reactor is reserved for those reactors in which the production of plutonium-239 (or uranium-233) from fertile materials is greater than the amount of fissile material consumed in the reactor. The ratio of the number of fissile atoms produced to that consumed is called the "breeding ratio" or "conversion ratio." A reactor that is designed so that the breeding ratio can exceed one is called a "breeder reactor." When this happens, the fuel output is greater than the fuel input. This (potential) feature was one of the reasons that nuclear energy was often described as a magical energy source.

In commercial reactors now in operation around the world, like LWRs and HWRs, the breeding ratio is less than one; they are referred to as "converter reactors." Typically, a light water reactor converts just under two percent of the uranium-238 into plutonium isotopes, about two-thirds of which consists of the fissile isotopes plutonium-239 and plutonium-241, while the rest consists of the non-fissile isotopes, mainly plutonium-240. Almost half of this plutonium is consumed during normal reactor operation, leaving the rest in the spent fuel. The plutonium consumed during reactor operation typically contributes about one-fourth to one-third of the energy generated in light water reactors.[34]

Theoretically, it is possible to use breeder reactors to vastly increase the amount of fissile material available for future use while producing energy for current use. The amount of time required to double the quantity of fissile material is called the "doubling time." For breeder reactors that convert uranium-238 into plutonium-239, theoretical doubling times are 9 to 16 years, depending on reactor design; for reactors that convert thorium-232 into uranium-233, doubling times are estimated at 91 to 112 years. A longer doubling time means that a larger resource base of relatively scarce uranium-235 would be required to create an extensive nuclear energy system.

Since doubling times for breeding U-233 are far longer than for breeding Pu-239, almost all breeder reactors so far have been built to breed Pu-239. A further disadvantage of thorium-232-based breeder reactors cycle is the high gamma radioactivity due to contaminants in recovered uranium-233. This radioactivity arises mainly from the decay products of uranium-232, which is created in thorium-uranium fueled breeders by various nuclear reactions.[35] India seems to be the only country with a substantial active program to pursue U-233 breeding, since it has very large thorium-232 reserves, which are far greater than its domestic uranium-238 resources.

The number of neutrons per fission required for successful operation of a breeder reactor is considerably greater than for a converter reactor. This is because in addition to the one neutron per fission required to maintain the nuclear chain reaction in the reactor, at least one more is required to convert one atom of U-238 into an atom Pu-239 in order to maintain a breeding ratio of one or more. In practice, since some neutrons are absorbed

by the moderator, by other materials in the reactor vessel, and by the reactor vessel itself, the number of neutrons required for a breeding ratio greater than one is considerably more than two per fission.

The number of neutrons produced per fission from U-235 or Pu-239 when fissioned by slow (thermal) neutrons is 2.07 and 2.14 respectively; neither of these ratios is sufficiently large to permit the breeding ratio to be greater than one. In other words, there are not enough neutrons available to produce enough plutonium so it will exceed the fissile materials consumed and simultaneously maintain the chain reaction, given other neutron loss mechanisms.

To overcome this problem, breeder reactor designers take advantage of the fact that if the nuclei of U-235 or Pu-239 are bombarded by fast neutrons (energies of several hundred KeV or more), then the number of neutrons per fission increases substantially. For instance, the number of neutrons per fission for 5 MeV neutrons rises to about 3 for U-235 and to about 3.5 for Pu-239. Pu-239 breeder reactors employ this property by using fast neutrons to accomplish both fuel breeding and energy production. Breeder reactors using fast neutrons are also called "fast breeders" or "fast neutron reactors."

Fast breeders, by definition, need no moderators which slow down neutrons, since they use fast neutrons for fission and breeding. They cannot use ordinary water or heavy water as a coolant because these materials also act as moderators. Gases, which have low density, or atoms with heavy nuclei (mass numbers much greater than one), such as sodium metal, can be used as coolants in fast breeders. Molten salt has also been proposed. Liquid sodium, which has a mass number of 23, compared to 1 for ordinary hydrogen and 2 for deuterium, is the most common breeder reactor coolant. Since a coolant must continually flow across fuel elements, it must be a gas or liquid. Since sodium is a solid at room temperature, it must be maintained in liquid form in a breeder reactor by heating it continually, even when the reactor is shut down.

The most common type of breeder reactor is called the Liquid Metal Fast Breeder Reactor (LMFBR). Figure 5 [not available in on-line version of report] shows a schematic diagram of an LMFBR. A more recent variant of the liquid metal fast reactor design was being developed by Argonne National Laboratory until it was canceled in 1994. It was called the Integral Fast Reactor (IFR). This design had an electrolytic reprocessing plant that accompanied it. Electrolytic reprocessing, called electrometallurgical processing or pyroprocessing, is still being pursued by the DOE at Argonne West in Idaho.[36]

Sodium catches fire on contact with air and explodes on contact with water. Further, the nucleus of ordinary sodium absorbs a neutron and turns into a highly radioactive isotope sodium-24. This is a major threat in case of a breeder reactor accident. To prevent leakage of sodium-24 into the environment, sodium-cooled reactors are designed with two liquid sodium loops. The secondary, non-radioactive sodium loop draws heat from the primary loop and, in turn, is used to boil water in a steam generator. The December 1995 accident at the Japanese breeder reactor at Monju involved a large leak of sodium from the secondary loop.

Despite its theoretical attractiveness in converting non-fissile into fissile material, the breeder reactor has turned out to be a far tougher technology than thermal reactors. Despite five decades of effort during which many pilot and "demonstration" plants have been built, the sodium-cooled breeder reactor design remains on the margin of commercial nuclear technology. The magic of fuel multiplication has not yet been realized on any meaningful scale relative to nuclear electricity generation levels. Plutonium can also be mixed with uranium for use in thermal reactors. Generally, both plutonium and uranium are mixed after conversion into a dioxide chemical form. For this reason, the plutonium-uranium fuel mixture is called "mixed oxide" fuel, or "MOX" fuel for short.

The "Nuclear Fuel Cycle"

Nuclear power as initially conceived was to be based on using both the natural fissile material uranium-235 and increasing the amount of fissile material by converting uranium-238 (or thorium-232) into fissile materials. In this scheme of things, uranium mining and milling would eventually be a supplement to the creation of fissile materials from an initial stock of fertile uranium-238 and thorium-232 in nuclear reactors.

Reprocessing plants would separate the fissile isotopes from the spent fuel for use in fuel fabrication plants. Many of the long-lived highly radioactive fission products resulting from power generation would be used for a variety of purposes, ranging from nuclear medicine to food irradiation to thermoelectric generators to a vast array of science fiction type of applications that became the subject of much swooning prose in the decade that followed the end of World War II. There would be little waste. There would be a nuclear fuel cycle.

However, it was recognized even in the early years that large scale use of nuclear energy would produce fission products in such huge quantities that some arrangements would have to be made for their disposal. But expectations that disposal in salt mines would be a relatively straightforward matter proved too optimistic, like so many other prognostications regarding nuclear power. (See Chapter 6.)(Not available on-line.)

To complicate matters further, reprocessing and fabrication of plutonium into reactor fuel (whether for breeder reactors or light water reactors) turned out to be very expensive, while uranium resources were far more plentiful than anticipated in the 1950s. This made the use of plutonium as a fuel uneconomical, leading to a build-up of spent fuel (which is irradiated fuel discharged from a reactor) at power plant sites. The mounting plutonium stocks, both separated and in spent fuel, are a major source of concern as regards their proliferation potential.

Endnotes

28. Lamarsh 1983, p. 119.

29. Reactors that use thorium-232 as the raw material to produce fissile uranium-233 are also possible, but no significant commercial reactors of this type have been built.

30. See TMI Commission 1979 for an account of the accident.

31 See Lamarsh 1983, pp. 280-285.

32. Reactor control in water moderated and cooled reactors can also be accomplished chemically by adding a neutron absorbing material, generally boric acid, to the water. This kind of control is called chemical shim. It is not used by itself, but to supplement the control achieved by use of control rods.

33. Lamarsh 1983, p. 286. Reactivity relative to the fraction of delayed neutrons is measured in "dollars" and "cents." One dollar of reactivity occurs when the reactivity is equal to the proportion of delayed neutrons, at which stage the reactor is prompt critical. Evidently, to control the reactor, the reactivity must be kept below one dollar, which is why reactivity for normal reactor operation is measured in cents, with one cent being one-hundredth of the reactivity at prompt criticality.

34. This estimate is calculated as follows: With 3.3 percent enriched uranium fuel, after 30,000 megawatt days of burn-up, the spent fuel contains about 3.3 percent fission products and about 1 percent uranium-235. The energy release per fission for uranium-235 and plutonium-239 is about the same. Since about 1 out of every 3.3 fissions is plutonium (the rest being uranium-235), about 1/3.3, or 30 percent of the energy comes from plutonium. The fraction of energy from plutonium will vary with fuel enrichment and burn-up. Relative abundance data are from Benedict et al. 1981, Figure 3.3, p. 88.

35. Benedict et al. 1981, p. 378.

36. Sachs 1995, p. 33.

Glossary

^{0}C	Degree centigrade
absorbed dose	The amount of energy deposited in a unit weight of biological tissue. The units of absorbed dose are rad and gray.
actinides	The 14 elements following actinium in the Periodic Table.
activation product	An element that is transmuted from a non-radioactive into a radioactive material when its nucleus absorbs an elementary particle, such as a neutron.
AEC	United States Atomic Energy Commission, 1947-1974. Broken up in 1974 into the Energy Research and Development Administration (ERDA) and the Nuclear Regulatory Commission (NRC). ERDA later became the Department of Energy (DOE).
ALMR	Advanced liquid metal reactor.
alpha decay	The emission of a nucleus of a helium atom from the nucleus of an element, generally of a heavy element, in the process of its radioactive decay.
alpha particle	The nuclei of a helium atom (with two neutrons and two protons each) that are discharged by radioactive decay of many heavy elements, such as uranium-238 and plutonium-239.
alpha radiation	Radiation consisting of helium nuclei (atomic weight 4, atomic number 2) that are discharged by radioactive disintegration of some heavy elements, including uranium-238, radium-226, and plutonium-239.
Am	Americium, the next element after plutonium; atomic number 95.
AmO_2	Americium dioxide.
ANL	Argonne National Laboratory (USA).
APT	Accelerator Production of Tritium.
ATW	Accelerator driven Transmutation of Waste. A project of Los Alamos National Laboratory (USA).

Atomic number (symolized Z)	The number of protons in nucleus. It determines the chemical properties of an element.
atomic weight	The nominal atomic weight of an isotope is given by the sum of the number of neutrons and protons in each nucleus. The exact atomic weight differs fractionally from that whole number, because neutrons are slightly heavier than protons and the mass of the nucleus is also affected by the binding energy.
becquerel	A unit of radioactivity equal to one disintegration per second. It is an extremely small unit, equal to about 27 picocuries.
beta decay	The emission of electrons or positrons (particles identical to electrons, but with a positive electrical charge) from the nucleus of an element in the process of radioactive decay of the element.
beta particle	Electrons or positrons (positively charged electrons) emitted by many elements in the course of radioactive decay.
beta radiation	Radiation consisting of electrons or positrons emitted in many radioactive disintegrations, at speeds approaching the speed of light.
Bi	Bismuth. Atomic number 83.
binding energy	The energy that is required to separate the nucleons in a nucleus into separate, free particles.
blanket	The fuel and transmutation assemblies that make up the subcritical system surrounding the central target area of a transmuter.
BNFL	British Nuclear Fuels plc (UK).
Bq	Becquerel, a S.I. unit of radioactivity, one disintegration per second.
breeder reactor	A reactor that is designed to produce more fissile material than it consumes; also sometimes called "fast reactor" since most breeder reactors use fast neutrons for sustaining the nuclear chain reaction.
Btu	British thermal unit -- the amount of energy gained by a pound of water when its temperature is increased by one degree Fahrenheit.
BWR	Boiling Water Reactor - a light water reactor that boils the reactor

	coolant. The resultant steam is used to drive a steam turbine.
calorie	A unit of heat or energy sufficient to raise the temperature of 1 gram of water by 1 degree Celsius. In dietetics, the kilocalorie is the unit usually used, frequently called a "calorie," omitting the prefix.
CANDU reactor	CANada Deuterium Uranium reactor, a heavy water moderated power reactor used in Canada.
CAPRA	“Consommation Accrue de Plutonium en Réacteur rApide.”
Cd	Cadmium. Atomic number 48.
CEA	Commissariat à l’Énergie Atomique (France).
CERN	European Organization for Nuclear Research.
chemical shim	Control of the chain reaction in a nuclear reactor by controlling the chemical composition of coolant water; used as a supplement to the employment of control rods.
Cm	Curium; the next (artificial) element after americium. Atomic number 96.
CMPO	N-octyl-phenyl-di-isobutyl-carbamoylmethyl-phosphine-oxide; an organic solvent extraction compound used for TRUEX process.
COGEMA	Compaignie Générale des Matières Nucléaires (France).
cohort	A group of individuals having a statistical factor (such as age) in common in a demographic or epidemiological study.
control rods	Rods made out of a neutron absorbing material that enable control of the chain reaction in a nuclear reactor.
coolant	Fluid circulated through a reactor to transfer heat from the fuel to its destination.
cooling	A period of delay following discharge of fuel from a reactor, allowing much of the initial radioactivity to decay.
core	The region of a nuclear reactor in which a chain reaction can take place.
critical mass	The amount of a fissile substance that will allow a self-

	sustaining chain reaction. The amount depends both on the properties of the fissile element and on the shape of the mass.
criticality	A fission chain reaction proceeding at a steady or increasing rate. In a reactor, the normal operating condition; elsewhere an accident to be strictly avoided.
cross-section	The probability of interaction, for instance between a nucleus and a neutron flux, measured in barns (10^{-24} cm^2).
curie	Unit of radioactivity equal to the radioactivity of 1 gram of radium -226. It is equal to 37 billion disintegrations per second.
decay	See radioactive decay.
decay correction	The amount by which the calculated radioactivity (for example, of a release of radioisotopes) must be reduced after a period of time, to allow for its radioactive decay during that time.
decay heat	Energy released by the radioactivity of fission products in reactor fuel after fission has ceased.
decommissioning	Decontamination and dismantlement of retired, contaminated facilities and removal and/or disposal of the resulting wastes.
delayed neutrons	Neutrons that are not emitted promptly after a fission reaction but rather after a delay.
depleted uranium	A by-product of uranium enrichment, the most common chemical form of which is depleted uranium hexafluoride (DUF_6). Natural uranium is composed of three isotopes: uranium-238 (99.284 percent); uranium-235 (0.711 percent); and uranium-234 (0.005 percent), all of which are radioactive. The purpose of uranium enrichment is to concentrate uranium-235, the fissile isotope, in one stream. The other stream which is low in uranium-235, is called "depleted uranium," which contains about 0.2 to 0.3 percent uranium-235.
deuterium	An isotope of hydrogen with atomic mass of two, having one proton and one neutron in the nucleus; non-radioactive.
DIAMEX	DIAMide Extraction; a typical process flowsheet based on diamide extractants for minor actinide separation.
DIDPA	Di-isodecylphosphoric acid; an organic extractant.

Direct disposal	The consignment of discharged fuel in its entirety (apart perhaps from appendages) to a permanent repository. Contrast reprocessing.
DMDBTDMA	Di-methyl-di-butyltetradecylmalonamide; a diamide-type extractant.
DOE	United States Department of Energy, created in 1977 by the elevation of ERDA to cabinet status.
doppler coefficient	In a reactor, the effect of temperature on the probability of fission in a reactor. A negative value is important for stable operation.
dose limit	Regulatory limit set on the amount of radiation that an individual may receive from artificial sources (excluding medical sources). Worker limits are set higher than general population limits.
dose reconstruction	Estimating exposure by considering emissions, environmental measurements, and routes of exposure.
DTPA	Diethylenetriaminopentaacetic acid.
dual-purpose reactor	A reactor that produces steam for energy use as well as tritium and/or plutonium for military use.
DWPF	Defense Waste Processing Facility, the name of the vitrification plant for high-level radioactive wastes at the Savannah River Site.
EA	Energy Amplifier.
EBR II	Experimental Breeder Reactor II, in Idaho.
ECU	European Currency Unit.
EFR	European Fast Reactor.

EFTTRA	Experimental Feasibility of Targets for Transmutation; an international collaboration among Commissariat à l'Énergie Atomique (CEA), Netherlands Energy Research Foundation (ECN), Électricité de France (EDF), Forschungszentrum Karlsruhe (FZK), Institute for Advanced Materials, The Netherlands (IAM), and Institute for Transuranium Elements (ITU).
Electron	An elementary particle carrying 1 unit of negative electric charge. Its mass is 1/1837 that of a proton.
electron-volt	A measure of energy used for atomic phenomena (abbreviation eV). It is the amount of energy acquired by an electron traveling through one volt of electric potential difference. It is equal to $1.6*10^{-19}$ joules.
electrolysis	Process depending on the passage of electric current through a conducting "electrolyte."
enrichment	An artificial increase in the proportion of one isotope of an element (usually uranium) by partial separation from others, leaving depleted "tails."
ERDA	United States Energy Research and Development Administration, created in 1974 from the break up of the AEC.
eV	Electron-volt
exa-	Prefix for one million trillion (or 10^{18}). One metric ton of U.S. coal on the average is approximately 25 billion joules. Therefore one exajoule is equivalent to about 40 million metric tons of U.S. coal.
external radiation dose	The dose from sources of radiation located outside the body. This is most often from gamma rays, though beta rays can contribute to dose in the skin and other relatively superficial tissues.
extractant	The effective component in a solvent extraction process.
fast breeder reactor	A fast neutron reactor that generates more fissile material than it uses.
fast neutron reactor	A reactor that uses fast neutrons to sustain the chain reaction.

fertile material	A material that is not fissile, but which can be converted into a fissile material; uranium-238 and thorium-232 are the principal fertile materials.
fissile	Capable of undergoing nuclear fission induced by thermal neutrons, as distinct from "fissionable"-- subject to fission only when induced by higher energy neutrons.
fissile material	A material whose nucleus can be fissioned when it absorbs a low energy (ideally zero energy) neutron. Fissile materials can sustain chain reactions. . Well-known examples are plutonium-239 and uranium-235.
fission	The splitting of the nucleus of an element into fragments. Heavy elements such as uranium or plutonium release energy when fissioned.
fission product	Any isotope created by the fission of a heavy element. Fission products are usually radioactive.
fissionable material	A material that can undergo nuclear fission when bombarded by a neutron. Some materials like uranium-238 are fissionable because they undergo fission when bombarded by energetic neutrons, but they are not fissile.
FP	Fission product.
fusion	The combining of two nuclei to form a heavier one. Fusion of the isotopes of light elements such as hydrogen or lithium gives a large release of energy.
gamma radiation	Electromagnetic waves released during radioactive decay that can ionize atoms and split chemical bonds. Gamma rays are similar to X-rays, the latter term being applied usually to electromagnetic waves generated by electron accelerators, as for instance in medical equipment.
GeV	Giga-electron volt (1 GeV = 10^9 eV).
giga-	Prefix for billion (or 10^9) "Billion" corresponds to "milliard" in France.

gigawatt	One billion watts; the approximate electrical capacity of a large nuclear power plant.
gray	A unit of absorbed radiation dose equal to 100 rads.
GWd	Giga-watt day.
GWd/tHM	A measure of irradiation of fuel in a reactor corresponding to the gross thermal energy obtained from a metric ton of the fuel expressed in units of gigawatt days thermal per metric ton of heavy metal.
GWe	Giga-watt electric.
Gy	Gray.
half-life	The amount of time that it takes half of a given quantity of a radioactive element to decay.
heavy water	Water in which deuterium has replaced ordinary hydrogen; the symbol D is often used for deuterium. The chemical formula for ordinary water is H_2O; that for heavy water is D_2O.
HEU	Highly enriched uranium.
HLLW	High level liquid waste resulting from fuel reprocessing operations.
HLW	High level waste in solidified form.
HTGR	High-Temperature Gas-Cooled Reactor.
I	Iodine. Atomic number 53.
IFR	Integral Fast Reactor, a variant of the liquid metal fast breeder reactor design.
induced radioactivity	Radioactivity produced in any material as a result of nuclear reactions, especially by absorption of neutrons.
internal radiation dose	The dose to organs of the body from radioactive materials inside the body. It may consist of any combination of alpha, beta, and gamma radiation.
ionize	To split off one or more electrons from an atom, thus leaving it

	with a positive electric charge. The electrons usually attach to other atoms or molecules giving them a negative charge.
isotope	Atoms of the same element that have the same number of protons (and hence the same chemical properties), but a different number of neutrons, and therefore, different atomic weights.
JAERI	Japan Atomic Energy Research Institute.
JOYO	An experimental fast reactor (Japan).
joule	A metric unit of energy, equal to one watt of power operating for one second; one kilowatt-hour is equivalent to 3.6 million joules.
k_{eff}	Effective neutron multiplication factor.
kilo-	Prefix for one thousand
kiloton (KT)	In the context of nuclear weapons, this term, which means 1,000 tons, is always used as a measure of explosive power. It is equal to the explosive power of 1,000 tons of TNT.
kilowatt	One thousand watts, a common measure for electrical power capacity.
kilowatt-hour	A unit of energy equal to 3.6 million joules. It is the amount of energy contained in a one-kilowatt source operating for one hour. Abbreviation: kWh. When used in reference to electrical energy, the suffix, "e", for electrical, is often attached, making the abbreviation kWhe. It is common in electrical engineering practice to omit the “e”. When used in reference to thermal, or heat energy, the suffix "t" for thermal is generally attached, making the abbreviation kWht. In this report, the abbreviation kWh refers to electrical kilowatt hours. Thermal energy is expressed in joules or kWht.
kWhe	Kilowatt-hour electrical.
kWht	Kilowatt-hour thermal, equal to 3.6 million joules of thermal (heat) energy. The specification of energy as thermal or electrical is important in electrical generation because only a portion of thermal energy can be converted to electricity.
LANL	Los Alamos National Laboratory (USA).
LANSCE	Los Alamos Neutron Science Center.

LBE	Lead-bismuth eutectic. This eutectic mixture (that is, a specific mixture of two materials that yields the lowest, common melting point) has been used in Russian nuclear submarine reactor design.
LEU	Low enriched uranium.
light water reactor	The most common type of nuclear reactor in the world. Uses light water (ordinary water) as a moderator (to slow down neutrons in the reactor) and a coolant. Light water reactors are built in two variants: pressurized water reactors and boiling water reactors.
linear energy transfer (LET)	Refers to the rate of energy transfer (and thus damage) per unit at distance traveled. For example, alpha is high-LET radiation, while photons and electrons are low-LET radiation.
LMFBR	Liquid Metal Fast Breeder Reactor.
low-level radioactive waste	A catch-all category of waste defined by U.S. law as all wastes that are not in other categories such as "high-level" waste and mill tailings; radioactivity of "low-level" wastes varies widely and includes both short- and long-lived isotopes.
LWR	Light Water Reactor, a reactor that uses ordinary water, H_2O, as the moderator and coolant; comes in two basic variants, the BWR and the PWR.
mass number (symbolized A)	The sum of the number of protons and the number of neutrons in a nucleus.
mega-	Prefix for one million (or 10^6))
megawatt	One million watts, a common measure of generating capacity for large power plants. When used by itself in the context of electrical generation, it generally refers to electrical generating capacity, and is abbreviated as MW or MWe. The rate of heat generation can also be measured in megawatts, in which case the term megawatts thermal is used, abbreviated as MWt or MWth.
megawatt-days	The amount of energy generated by one megawatt of power output over one day. This is used to measure the degree of burn-up of nuclear fuel, and generally refers to thermal energy output extracted from the fuel.

meltdown	The accidental melting of nuclear reactor fuel rods and fuel.
metric ton	1,000 kilograms; approximately 2,20 pounds, and very nearly equal to a British ton (2,240 pounds). The usual U.S. ton measurement, called a short ton, is 2,000 pounds.
MHTGR	Modular High-Temperature Gas-Cooled Reactor.
micron	One millionth of a meter (or 10^{-6}m or μ[mu])
mill	One-tenth of one U.S. cent. The cost of electrical power is often expressed in terms of mills per kilowatt hour.
mill tailings	A slurry of about 40 percent solids (including radioactive particles and chemically hazardous metals) and 60 percent liquid, primarily water.
minor actinides	Transuranic radionuclides, other than plutonium, that pose significant waste management issues. They are: neptunium, americium, and curium.
moderation ratio	Ratio of moderator volume to fuel volume. Typical value for a pressurized water reactor is 1.7. Undermoderation indicates lower values of moderation ratio (range 1-1.5); overmoderation indicates higher values (range 2-3).
moderator	A material used in a nuclear reactor to slow down the fast neutrons emitted in the process of fission.
MOX	Mixed oxide fuel. A fuel composed of a mixture of plutonium dioxide and uranium dioxide.
MRS	Monitored Retrievable Storage, a centralized storage facility for spent fuel from nuclear reactors.
multiplication factor	The number of fission reactions on average caused by a single fission. A multiplication factor greater than one means a reactor is supercritical, equal to one means exactly critical, and less than one means subcritical.
MW	Megawatt (or 10^6 W).
MWd	Megawatt day (or 1 MWd = 10^6Wd)
MWe	Megawatt electrical, a measure of electrical generating capacity;

	also written as MW.
MWt	Megawatt thermal, a measure of the heat energy generated in a boiler or reactor; also written as MWth.
NEA	Nuclear Energy Agency of the Organisation of Economic Co-operation and Development.
neutron	A neutral elementary particle that occurs in the nuclei of elements (except ordinary hydrogen); free neutrons decay into a proton, an electron and a neutrino. A neutron is about 1,838 times heavier than an electron.
neutron capture	The capture by the nucleus of an element of a neutron initially external to it.
neutron flux	The number of neutrons crossing a unit area per unit time.
neutron spectrum	The energy distribution in the neutron flux of a reactor.
NpO_2	Neptunium dioxide.
NPT	The nuclear Non-Proliferation Treaty.
NRC	United States Nuclear Regulatory Commission, formed in 1974 from the breakup of the Atomic Energy Commission.
NRC-NAS	National Research Council of the National Academy of Sciences, administered jointly by the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.
nuclear fission	The splitting of the nucleus of a heavy element into two lighter nuclei, generally accompanied by the release of one or more neutrons and energy.
nuclear fusion	The fusion of two light nuclei, accompanied by the creation of a new light nuclei and the release of energy.
nucleon	Proton or neutron occurring in the nucleus of an element.
nucleus	The nucleus of an atom is the central core that comprises almost all the weight of the atom. All atomic nuclei (except H-1, which has a single proton) contain both protons and neutrons.
nuclide	A particular isotope.

OMEGA	Options Making Extra Gains from Actinides and Fission products; Japanese
pathway analysis	An analysis of the ways in which toxic or radioactive substances can reach human beings from a factory, place, or process in which they are made, used, stored or dumped via air, water, soil, the food chain, or some combination of these pathways.
peta-	Prefix for one thousand trillion (or 10^{15}). Energy use on a large scale is often measured in petajoules. One metric ton of U.S. coal on the average is approximately 25 billion joules. Therefore one petajoule is equivalent to about 40,000 metric tons of U.S. coal.
photon	The indivisible unit, or quantum, of electro-magnetic radiation. The energy of the photons determines the nature of the radiation, from radio waves at the lowest energy levels, up through infra-red, visible, and ultra-violet light, to X-or gamma-rays, which have energy high enough to ionize atoms.
pin (fuel)	A tube packed with fuel pellets,used in suitably spaced groups of up to several hundred, variously termed clusters, elements, sub-assemblies, etc.
plutonium	A highly toxic, heavy, radioactive metallic element. There are 15 isotopes of plutonium, of which only five are produced in significant quantities: plutonium-238, -239, -240, -241, and -242. Plutonium-239 is the most important plutonium isotope as it is fissile and is used in nuclear weapons and some reactors. On the other hand, plutonium-240 is unsuitable for use in nuclear weapons and reactor fuel. Thus, in a reactor whose main purpose is plutonium production, the rate at which plutonium-240 is formed controls the length of time fuel is allowed to remain under irradiation. Plutonium is categorized according to plutonium-240 content, as follows: super-grade has 2-3% Pu-240; weapons-grade has less than 7% Pu-240; fuel-grade has 7-18 (or sometimes given as 7-19) % Pu-240; and reactor-grade has 18 or greater (or 19 or greater) % Pu-240. (Note: Despite what the name implies, "reactor-grade" plutonium has been used successfully to make a nuclear bomb.) Atomic number 94.
positron	An elementary particle with a positive electric charge, but in other respects identical with an electron.
prompt critical	The condition of becoming critical with prompt neutrons only.
prompt neutrons	Neutrons emitted concomitantly with a fission reaction.

proton	An elementary particle with a positive charge equal to that of an electron, but which is about 1,836 times heavier than an electron. It is given the value 1 on the scale of atomic weights.
PuO_2	Plutonium dioxide.
PUREX	A most commonly used process flowsheet based on TBP for fuel reprocessing.
PWR	Pressurized Water Reactor, a light water reactor that has water under high pressure (primary water) in the reactor which serves as a moderator and coolant. This primary water heats up water in a secondary circuit. Only the water in the secondary circuit is converted to steam, while the primary coolant remains in liquid form.
rad	A unit of absorbed radiation dose defined as deposition of 100 ergs of energy per gram of tissue. One erg is one-ten-millionth part of a joule (one erg = 10^{-7} joules). A rad amounts to approximately one ionization per cubic micron.
radioactivity	The spontaneous discharge of radiation from atomic nuclei. This is usually in the form of beta or alpha radiation, together with gamma radiation. Beta or alpha emission results in transformation of the atom into a different element, changing the atomic number by +1 or -2 respectively.
radionuclide	Any radioactive isotope.
radiotoxicity	A number indicating the potential of a radionuclide to cause health damage (cancer). It is often estimated by calculating the amount of water or air needed to dilute the pure substance so that the solution corresponds to the regulatory drinking water or air concentration limits. The radiotoxicity index does not account for whether the specific radionuclide will in fact reach the target population and result in an exposure.

reactivity	A number that measures whether and by how much a reactor is subcritical or supercritical. A reactivity of zero corresponds to a reactor being exactly critical. Reactivity greater than zero means the reactor is supercritical, while a reactivity less than zero indicates it is subcritical.
reactor core	The core of a reactor consists of the fuel, moderator (in the case of thermal reactors) and coolant.
relative biological effectiveness (RBE)	A factor that can be determined for different types of ionizing radiation, representing the relative amount of biological change caused by 1 rad. It depends upon the density of ionization along the tracks of the ionizing particles, being highest for the heavy particles: alpha rays and neutrons.
relative risk	The ratio of disease incidence (or mortality) in an exposed population to that in an unexposed population.
rem	Radiation dose (in rad) multiplied by an empirical factor, called quality factor, which represents the biological effectiveness of a particular kind of radiation to cause biological damage relative to gamma radiation. The dose in rems is the dose in rads multiplied by the quality factor.
reprocessing	The chemical separation of irradiated nuclear fuel into uranium, plutonium, and fission products.
roentgen	A unit of gamma radiation measured by the number of ionizations it causes in air. In non-bony biological tissue one roentgen is, for practical purposes, approximately equal to one rad.
Rubbiatron	A specific accelerator transmutation system named after Italian physicist Carlo Rubbia.
σ_f ("sigma sub f")	Fission cross-sections.
σ_c ("sigma sub c")	Capture cross-sections.
SESAME	“Séparation Extraction Sélective de l’Américium par Moyens Électrochimiques” (Selective Extracting Separation of Americium by Means of Electrolysis).
shielding	Material used to absorb radiation before it can cause damage or injury.
sievert	The S.I. unit of equivalent absorbed radiation dose equal to 100 rems.

SNS	Spallation neutron source.
solubility	The ability to dissolve in water. For instance, the less soluble a given amount of material, the more difficult it is for the body to remove it. An insoluble material inhaled into the lungs for example would have more time to do damage to the lungs.
source term	The amount of a specific pollutant emitted or discharged to a particular medium, such as the air or water, from a particular source.
spallation	A high energy nuclear reaction in which an elementary particle collides with a target made of a heavy material. The nucleus of the target, when struck, emits a number of particles. The word spallation comes from the word "spall," which means "to chip off" because the emitted particles are essentially chipped off of the original heavy nucleus. In the context of transmutation, spallation refers to accelerator based schemes in which a heavy target, such as lead, is struck by an accelerated proton and emits neutrons. These neutrons act as a supplemental neutron source for the reactor.
specific activity	A measure of the radioactivity of a unit weight (generally one gram) of material.
spontaneous fission	The spontaneous splitting of the nucleus into two new nuclei, generally with the emission of one or more neutrons and the release of energy.
stripping	The extraction (or "back-extraction") of a material from the solvent that has been used to extract it (along with other materials) from an aqueous solution.
sub-critical reactor	A nuclear reactor that is configured to operate with an external source of neutrons to supplement internally generated neutrons to maintain the chain reaction.
SUPERFACT	Actinide incineration experiment in fast reactor, Phénix (France)
Sv	Sievert.

target	An assembly suitable for absorbing neutrons in a reactor that contains radionuclides to be transmuted. Generally, the targets contain radionuclides that do not form the primary fuel for the reactor (e.g. fission products or, sometimes, minor actinides).
TBP	Tributylphosphate; an organic extractant used for PUREX process.
Tc	Technetium. Atomic number 43.
thermal reactor	A reactor that uses thermal (or slow) neutrons to sustain the chain reaction
thermonuclear weapon	A nuclear weapon that gets a large part of its explosive power from fusion reactions.
tHM	Metric ton of heavy metal.
TNT equivalent	The weight of TNT which would release the same amount of energy as a particular nuclear explosion. One ton of TNT releases approximately 1.2 billion calories (that is, 5.1 kilojoules per gram). Nuclear explosions are usually measured in kilotons (KT) or megatons (MT).
ton	See metric ton.
transients	Surges or declines in reactors' parameters, such as power levels Or neutron flux, in a reactor, often referring to sudden changes in these parameters.
transmutation half-life	The amount of time it takes to transmute half of a long-lived radionuclide in a reactor.
transmuter	A nuclear reactor used for transmutation. It most commonly Refers to a subcritical reactor that uses transuranic elements as fissile material and is driven by accelerator-produced neutrons.
transuranic element	An element with atomic number greater than 92, which is the atomic number of uranium.
tritium	A radioactive isotope of hydrogen with a half-life of 12.3 years having one proton and two neutrons in its nucleus. Its principal use is in nuclear weapons.

TRU	Transuranic element; for example: Neptunium, Plutonium, Americium, Curium, Berkelium, Californium.
TRUEX	TRansUranium Extraction; a typical process flowsheet based on CMPO for minor actinide separation.
U	Uranium, the heaviest element occurring naturally in significant amounts; atomic number 92.
UN	Uranium nitride.
UO_2	Uranium dioxide.
UP3	A reprocessing plant in LaHague (France).
vitrification	The conversion of wastes to a glassy form for permanent disposal.
watt	A metric unit used to measure power, that is the rate of energy generation or consumption. One watt is equal to one joule per second. One horsepower is equal to 746 watts.
watt-hour	One watt of power operating for one hour; equivalent to 3,600 joules of energy.
WVDP	West Valley Demonstration Plant, the name of the vitrification plant for high-level radioactive wastes at West Valley, New York.
yield	The energy released by a nuclear explosion.
zircaloy	An alloy of zirconium with 1.2 to 1.7 percent tin and smaller quantities of iron, chromium, and nickel used for making the tubes into which the nuclear fuel for light water reactors is inserted.[464]
Zr	Zirconium. Atomic number 40.

[464] Benedict, Pigford, and Levy 1981, p. 323-324.

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